Author	Insttituion	Day	Number	Poster title
Brown, Douglas	New Mexico State University	Wednesday	1	Seeing the invisible with coherent x-ray diffraction imaging
Carnis, Jerome	Sogang University, Korea	Wednesday	2	Demonstration of feasibility of x-ray free electron laser studies of dynamics of nanoparticles in entangled polymer melts
Cha, Wonsuk	Materials Science Division, Argonne National Laboratory	Wednesday	3	Core-shell strain structure of zeolite microcrystals
Chen, Chun-Yu	NSRRC, Taiwan	Wednesday	4	Design of the coherent x-ray scattering beamline at the Taiwan Photon Source
Chen, Joe	University of Canterbury	Wednesday	5	Phase retrieval for intensity data averaged over different unit cells
Ekeberg, Tomas	Uppsala University	Wednesday	6	Three-dimensional imaging of viruses using LCLS
Gao, Yuan	Argonne National Laboratory	Wednesday	7	X-ray scattering from optically trapped nanoparticles
Gutt, Christian	University Siegen	Wednesday	8	Femtosecond speckle and coherence experiments at the CHG short-pulse facility DELTA
Hantke, Max	Uppsala University	Wednesday	9	High-throughput imaging of cell organelles with an x-ray laser
Huang, Xiaojing	National Synchrotron Light Source II	Wednesday	10	Dynamical artifacts in Bragg coherent diffraction imaging
Ilavsky, Jan	Argonne National Laboratory	Wednesday	11	Multiplexed coherent x-ray photon correlation spectroscopy measurements using micro-slit arrays fabricated by photolithography
Jacques, Vincent	CNRS/Laboratoire de Physique des Solides	Wednesday	12	Counting dislocations in micro-crystals with coherent x-rays: ex-situ and in-situ studies of the plastic deformation of InSb micro-pillars
Kimura, Takashi	Hokkaido university	Wednesday	13	Imaging of live cell in micro-liquid enclosure array by signle-shot x-ray free electron laser diffraction
Kwasniewski, P	DESY	Wednesday	14	Anomalous dynamics prevent the glass transition in a hard-sphere colloidal suspension
Letourneau, Pierre-David	Columbia University	Wednesday	15	Inverse problem theory for x-ray diffraction of nano-particles
Leyffer, Sven	Mathematics and Computer Scien	Wednesday	16	Optimization problems in phase retrieval
Li, Kenan	Northwestern University	Wednesday	17	Simulation of paraxial wave propagation for zone plate optics
Malm, Erik	Colorado State University	Wednesday	18	Signal amplification in coherent diffractive imaging using diffractive optics
Marathe, Shashidhara	Argonne National Laboratory	Wednesday	19	Single phase grating interferometers for probing x-ray beam coherence area
Monteforte, Marianne	University College London	Wednesday	20	Coherent diffractive imaging of FePt nanocrystals embedded in a matrix
Nashed, Youssef	Mathematics and Computer Science Division, Argonne National Laboratory	Wednesday	21	PtychoLib: parallel ptychographic reconstruction
Parsons, Aaron	Diamond Light Source	Wednesday	22	Current and future applications of state-of-the-art photon counting detectors at I13
Phillips, Nicholas	Imaging CoE	Wednesday	23	Characterising structural disorder in bionanocrystals at the synchrotron and x-ray free electron laser
Rasshchupkyna, Mariya	The Hamburg Centre for Ultrafast Imaging, Max Planck Institute for Intelligent Systems, Stuttgart	Wednesday	24	Statistical description of four-point angular correlations: the concept of dominant orientational modes
Rodriguez, Jose	UCLA	Wednesday	25	Three-dimensional coherent x-ray diffraction imaging of whole, frozen-hydrated cells
Sandy, Alec	Argonne National Laboratory	Wednesday	26	ANL and LBNL 1k frame store CCD detector applied to XPCS at the APS
Schwenke, Joerg	University College London / London Centre for Nanotechnology	Wednesday	27	X-ray ptychographic imaging of human nuclei and chromosomes using reciprocal space up-sampling
Shimomura, Kei	Osaka University	Wednesday	28	High-resolution multislice x-ray ptychography in combination with precession measurement
Singer, Andrej	UCSD	Wednesday	29	Non-equilibrium structural dynamics of nanoparticles in LiN1/2Mn3/2O4 cathode under operando conditions
Tripathi, Ashish	Argonne National Laboratory	Wednesday	30	Ptychographic overlap constraint errors and the limits of their numerical recovery using conjugate gradient descent methods
Vaxelaire, Nicolas	Columbia University in the city of New York	Wednesday	31	Challenges and capabilities of coherent Bragg Imaging with polycrystalline thin film during mechanical loading
Williams, Garth	SLAC National Accelerator Laboratory	Wednesday	32	A novel real-time beam monitor for coherent and ultra-fast imaging
Zontone, Federico	ESRF	Wednesday	33	Coherent X-ray diffractive imaging at the ESRF beamline ID10: possibilities and challenges

Author	Insttituion	Day	Number	Poster title
Zozulya, Alexey	DESY	Wednesday	34	Ptychographic coherent diffraction imaging of polystyrene colloidal crystals in the vicinity of melting transition
Cha, Wonsuk	Materials Science Division, Argonne National Laboratory	Thursday	1	Three-dimensional Bragg coherent diffraction imaging using polychromatic x-rays
Chalkidis, Stefanos-Horst	University College London	Thursday	2	A visible light setup for near-field ptychography
Daurer, Benedikt	Uppsala University	Thursday	3	Toward diffractive imaging with single molecules using x-ray lasers
Enders, Bjoern	Technical University Munich	Thursday	4	Ptypy - A Python package for ptychography
Giewekemeyer, Klaus	European XFEL GmbH	Thursday	5	Towards experimental 3D coherent diffraction imaging from a large series of noisy diffraction patterns
Guehrs, Erik	Technical University of Berlin, Institute for Optics and Atomic Physics	Thursday	6	Soft x-ray tomoholography: exploration of advanced reference structures
Gutt, Christian	University Siegen	Thursday	7	Single particle structure determination using cross correlation techniques
Huang, R	Institute of Chemistry, Academia Sinica	Thursday	8	Toward XFEL imaging of Au-labeled icosahedral virus
Inoue, Ichiro	The University of Tokyo	Thursday	9	Development and demonstration of visibility-based x-ray photon correlation spectroscopy
Jurek, Zoltan	Center for Free-Electron Laser Science, DESY	Thursday	10	XMDYN: modeling radiation damage of XFEL irradiated samples
Kremenak, Jesse	University of Missouri	Thursday	11	Forbidden reflections in x-ray crystal truncation rods: using surface reference waves to distinguish charge and vibrational asymmetry in silicon
Kuppili, Venkata Sree Charan	UCL london	Thursday	12	Optimization of x-ray ptychography using the Excalibur detector at I-13 coherence beam line at Diamond
Lal, Jyotsana	Argonne National Laboratory/ Boston University	Thursday	13	X-ray correlation spectroscopy from dewetting polymer bilayer films
Lehmkühler, Felix	DESY	Thursday	14	Higher order x-ray intenstiy correlations in amorphous materials
Lhermitte, Julien	McGill University	Thursday	15	Measuring flows and velocity gradients with XPCS
Li, Po-Nan	Academia Sinica	Thursday	16	Method to enhance the resolution of x-ray coherent diffraction imaging for non-crystalline bio-samples
Mancini, Giulia	École Polytechnique Fédérale de Lausanne (EPFL)	Thursday	17	Ultrafast diffractive imaging with coherent electron pulses: light-induced ordering phenomena of functionalized nanoparticles
Mendez, Derek	Stanford University	Thursday	18	Correlated x-ray scattering: how to extract high-resolution information from solution measurements
Pietrini, Alberto	Uppsala university	Thursday	19	Statistical hit finding for XFEL biosamples
Robisch, Anna-Lena	Georg-August-University Göttingen	Thursday	20	Simultaneous phase retrieval of object and probe for extended wavefronts
Sandberg, R	Los Alamos National Lab	Thursday	21	Ultrafast imaging of shocked material dynamics with x-ray free electron laser pulses
Sandy, Alec	Argonne National Laboratory	Thursday	22	X-ray photon correlation spectroscopy at the APS with the Dectris EIGER detector
Shapiro, David	ALS, Lawrence Berkeley Lab	Thursday	23	Nanosurveyor: a new instrument for nano-tomography at the Advanced Light Source
Shemilt, Laura	TU Berlin	Thursday	24	Soft x-ray ptychography using holographic encoding
Shinohara, Yuya	The University of Tokyo	Thursday	25	Dynamics of silica particles in swollen rubber
Sichel-Tissot, Rebecca	Materials Science, Argonne	Thursday	26	X-ray probes of local composition and structure near defects in InGaN multi-quantum wells
Stockmar, Marco	Technische Universität München	Thursday	27	2D and 3D nano-imaging using near-field ptychography
Vaxelaire, Nicolas	Columbia University in the city of New York	Thursday	28	Challenges and capabilities of coherent Bragg imaging with nanoparticles
Zontone, Federico	ESRF	Thursday	29	Opportunities and challenges with high-energy coherent x-rays

Seeing the Invisible with Coherent X-Ray Diffraction Imaging

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In order to obtain the displacement field and strain distribution within and in the vicinity of nanostructures from measured Coherent X-ray Diffraction(CXD) experiments in kinematic far-field condition, we are required to solve a set of non-linear and non-local equations. A standard approach to solving these equations, which utilizes only the object's real space support and the intensity distribution in the vicinity of a Bragg peak as a priori knowledge, is the HIO+ER-algorithm. Despite its success for a range of applications, reconstructions for the case of highly strained nanocrystals are likely to fail. Here we utilize an extended form of Relaxed averaged alternating reflection (RAAR) algorithm which allows taking advantage of additional a priori knowledge of the local object geometrical shape and remedies HIO+ER's stagnation. Our approach achieves significant improvements in CXDI data analysis for highly strained crystals and greatly reduces sensitivity to the reconstruction's initial guess. These benefits are demonstrated in the reconstruction of twinned gold nanocrystals.

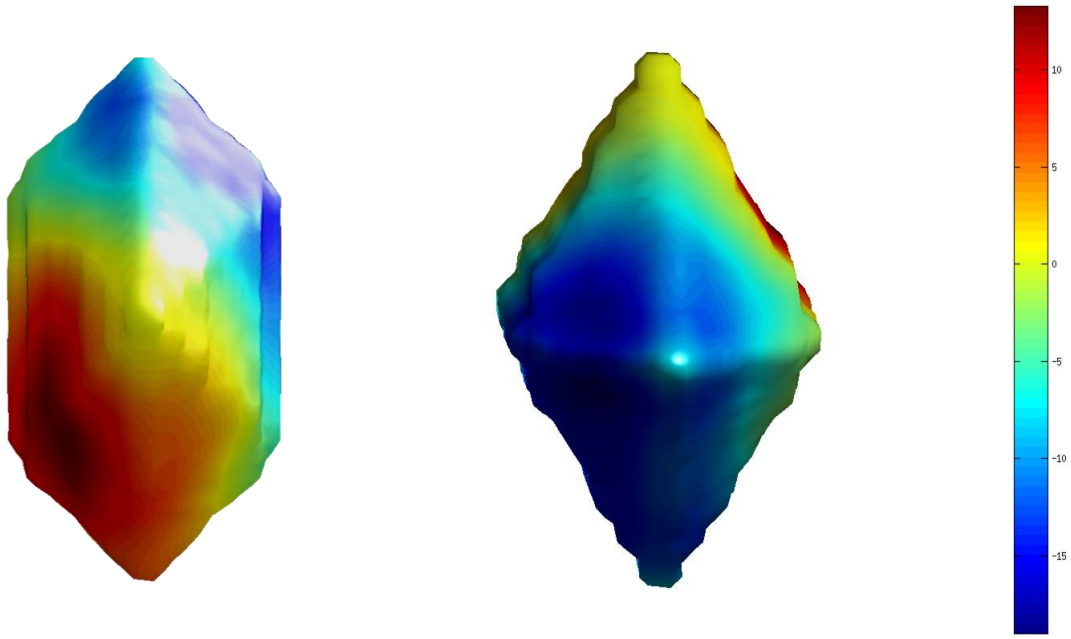

Figure 1: These are two views of a gold nanocrystal reconstructed using algorithms with MATLAB. This is a reconstruction of the shape and phase of the object. The internal strain is a unique benefit of CXDI, and is why reconstructing the particle this way is so valuable.

Demonstration of Feasibility of X-Ray Free Electron Laser Studies of Dynamics of Nanoparticles in Entangled Polymer Melts

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The recent advent of hard x-ray free electron lasers (XFELs) opens new areas of science due to their exceptional brightness, coherence, and time structure. In principle, such sources enable studies of dynamics of condensed matter systems over times ranging from femtoseconds to seconds. However, the studies of "slow" dynamics in polymeric materials still remain in question due to the characteristics of the XFEL beam and concerns about sample damage. Here we demonstrate the feasibility of measuring the relaxation dynamics of gold nanoparticles suspended in polymer melts using X-ray photon correlation spectroscopy (XPCS), while also monitoring eventual X-ray induced damage. In spite of inherently large pulse-to-pulse intensity and position variations of the XFEL beam, measurements can be realized at slow time scales. The X-ray induced damage and heating are less than initially expected for soft matter materials.

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Three-dimensional Bragg coherent diffraction imaging using polychromatic x-rays

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Coherent x-ray diffraction imaging (CXDI) has been developed to obtain three-dimensional images of various types of specimens at the nanoscale [1,2]. CXDI performed in the Bragg geometry has been employed to examine strain and deformation field distributions inside crystals [3,4]. There has been much effort to enhance the capability of the CXDI by seeking to overcome current spatial limitations and experimental constraints [5-6]. However the current state-of-art of this technique requires significant time to perform three-dimensional scans making it incompatible with most time-resolved studies.

In this study, we propose a new approach for CXDI with the goal of obtain three-dimensional images as well as internal strain distribution on short time scales using a polychromatic beam. Because polychromatic coherent x-rays produce multiple projections of coherent diffraction patterns at the same time, this approach takes advantage of rapid measurements without scanning samples. This innovative approach may provide opportunities for real time measurements and time-resolved three-dimensional imaging on isolated or extended crystalline samples. The measurements were performed at the 34ID-E beamline of the Advanced Photon Source, USA. The broadband coherent x-rays illuminated the sample and the coherent x-ray diffraction patterns were collected with an area detector. The setup for the three-dimensional Bragg coherent x-ray diffraction imaging will be discussed. The preliminary results will also be presented.

We thank the LDRD funding (#09269) for this project under the Argonne National Laboratory Hard X-ray Sciences Strategic Initiative.

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Core-Shell Strain Structure of Zeolite Microcrystals

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Zeolites are crystalline aluminosilicate minerals featuring a network of 0.3-1.5 nm wide pores, used in industry as catalysts for hydrocarbon interconversion, ion exchangers, molecular sieves, and adsorbents. For improved applications, it is highly useful to study the distribution of internal local strains because they sensitively affect the rates of adsorption and diffusion of guest molecules within zeolites. Here, we report the observation of an unusual "triangular" deformation field distribution in ZSM-5 zeolites by coherent x-ray diffraction imaging, showing the presence of a strain within the crystal arising from the heterogeneous core-shell structure, which is supported by finite element model calculation and confirmed by fluorescence measurement. The shell is composed of H-ZSM-5 with intrinsic negative thermal expansion while the core exhibits different thermal expansion behavior due to the presence of organic template residues, which usually remain when the starting materials are insufficiently calcined. Engineering such strain effects could have major impact on the design of future catalysts.

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A visible light setup for near-field ptychography

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Ptychography, originally suggested by Hegerl and Hoppe [1], is a method for solving the well-known phase problem whose implementation in coherent diffractive imaging techniques [2, 3] contributed significantly to the boost of the aforementioned field of science during the last decade. Using a scanning transmission microscopy setup, ptychography allows – in conjunction with redundant information from the overlap of the incident illumination between adjacent scan points [4] and powerful iterative algorithms [5, 6, 7] – for the simultaneous reconstruction of the object's transmission function and the scanning illumination profile. Recently, the applicability of this technique to near-field X-ray diffraction patterns was demonstrated [8], showing great promise as an alternative to other inline holography techniques. One benefit of near-field ptychography is the use of an illumination which extends almost completely over the reconstruction field of view, thus requiring fewer measurements and a less demanding dynamic range on the detector side. Furthermore, there is no need for wavefront corrections since wavefront imperfections are explicitly required by the presented reconstruction approach. In fact, the circumstance that the incident illumination does not need to be known *a priori* offers a wide range of ways for modifying the scanning beam. These features make near-field ptychography a very robust technique and therefore ideal for combining with tomography. In order to further explore this technique, we have designed and built a visible light setup aimed at finding the ideal conditions for laser-based near-field ptychography and to benchmark the technique for X-ray applications. We present our assembly in full detail and show first measurements.

We acknowledge financial support through the European Research Council (ERC, starting grant "OptImaX").

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Design of the Coherent X-Ray Scattering Beamline at Taiwan Photon Source

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The coherent X-ray scattering (CXS) beamline is one of the phase I beamlines designed for the Taiwan Photon Source, a new 3 GeV ring under construction at National Synchrotron Radiation Research Center in Taiwan. By using a pair of 2 meters-long in-vacuum undulators with the period length of 22 mm, CXS beamline will provide a highly coherent beam with the flux of 10^{10} photons/sec (at 5.56 keV) for X-ray photon correlation spectroscopy and coherent X-ray diffraction imaging. It will also share a part of beamtime for the users doing small-angle X-ray scattering (SAXS) experiments. The operating photon energy is designed within the range of 5.56-20 keV. The beam spot at sample position can be switched between 1 μm and 10 μm in both horizontal and vertical directions with using X-ray focusing optics. The high resolution and low-vibration sample stage will be installed. The sample-to-detector distance can be varying in the range from 0.5 meters to 12 meters. Data storage system and high-performance computing system will be provided for the users doing on-site preliminary calculations. The public opening of CXS beamline will be in the beginning of 2016.

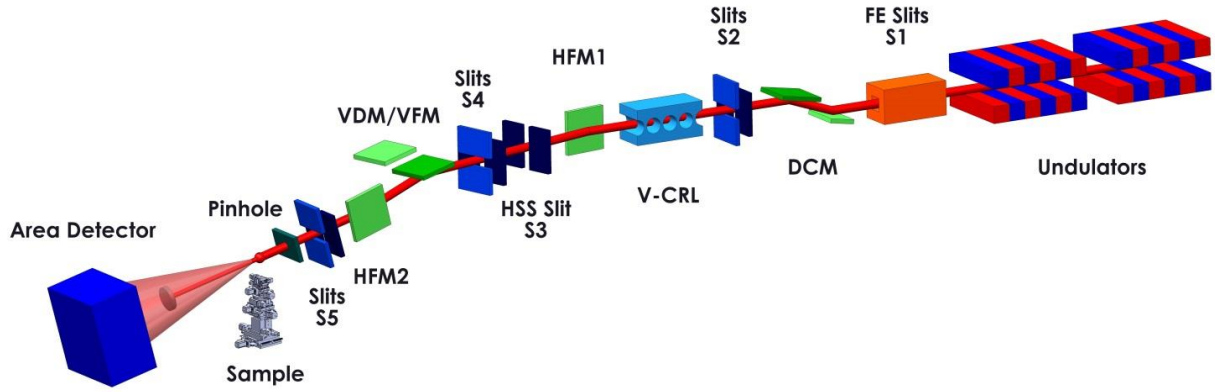

Figure 1: The optical layout of CXS beamline. (FE: front end; DCM: double crystal monochromator; V-CRL: vertical focusing compound refractive lens; HFM: horizontal focusing mirror; HSS: horizontal secondary source; VDM: vertical diffracting mirror; VFM: vertical focusing mirror)

Phase Retrieval for Intensity Data Averaged over Different Unit Cells

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In serial femtosecond nanocrystallography (SFX) using x-ray free-electron lasers, ensemble-averaged diffraction is measured from a collection of crystals of different sizes and shapes that each have a small number of unit cells [1]. These diffraction patterns contain information between the Bragg reflections which can be used to estimate the continuous diffracted amplitude of the contents of a single unit cell [2]. The problem is then analogous to that of reconstructing a single, non-periodic object (the contents of the unit cell in this case) from the amplitude of its Fourier transform, which is known to have a unique solution that can be found using iterative phase retrieval algorithms [3,4].

However, if there is more than one molecule in the unit cell, which is usually the case for protein crystals, then, as a result of the different terminations at the crystal surface, the diffraction from the crystal is not simply related to the diffraction from one unit cell [5]. To a first approximation, the diffraction is equal to the incoherent average over a set of unit cells that contain different molecular arrangements that are related to the space group at hand [5]. The data available are then the averaged intensity, constrained by the support of the unit cell and the symmetry operations that relate the different kinds of unit cell.

There are two approaches to solving the phase problem for this kind of data. The most straightforward is to use an iteration that involves reconstructing each unit cell using a simple reciprocal space projection, and then applying the symmetry relationships within and between the unit cells, as well as a support constraint in real space. A more elegant, and computationally less intensive, approach is to use an iteration that reconstructs the single asymmetric unit (the molecule itself) using a reciprocal space projection that incorporates the symmetry relationships [6]. The generality and applicability of these two approaches, and the corresponding reciprocal space projection operators, are investigated theoretically and by simulation.

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Toward diffractive imaging of single molecules using X-ray lasers

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The very short and intense pulses of recently developed X-ray lasers provide the possibility to study the structure of single biomolecules using coherent diffractive imaging techniques [1,2]. Both for large macromolecules and small viruses the measured signal intensity as of today is not sufficient to obtain structures from a single-shot experiment. However, there is great potential for a successful reconstruction of the three-dimensional structure by combining many randomly oriented single-shot diffraction patterns from a sample of reproducible particles and computationally solve the orientation problem prior to phase retrieval [3]. Along this path it is crucial to develop robust and automated procedures for a good pre-selection of patterns giving rise to a narrow size distribution from a large set of measured diffraction patterns (see Fig. 1). We therefore present computational methods for automated classification and size determination of small biological particles based on single-shot diffraction patterns with low intensity. The algorithms are tested on data measured at the Coherent X-ray Imaging (CXI) beamline at the Linac Coherent Light Source (LCLS) [4]. Being able to provide reliable information about the size of injected particles prior to orientation and reconstruction procedures is an important step toward the aim of solving the structure of single molecules with X-ray lasers.

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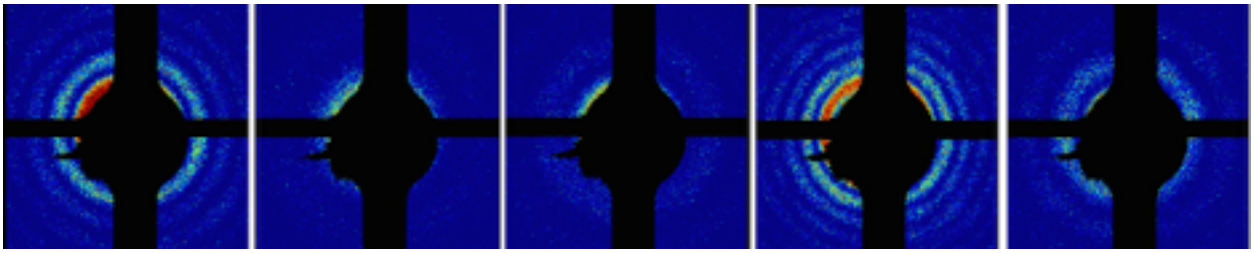

Figure 1: A selection of single-shot diffraction patterns of Omono river virus (OmRV) showing a variety of different sizes. The patterns were measured at CXI beamline (LCLS) with a CSPAD-140k detector [5] placed 2400 cm downstream of the interaction region at a photon energy of 7.1 keV.

Three-dimensional imaging of viruses using LCLS

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Free-electron lasers provide femtosecond X-ray pulses with a peak brilliance ten billion times higher than any previously available X-ray source [1]. Such a dramatic increase in a single experimental parameter is very unusual and often leads to far reaching implications in several areas of science. In structural biology, it has been suggested that such pulses will outrun key damage processes and allow structure determination without the need for crystallization [2]. So far applications have been limited to 2D projection images [3]. Here we present a proof-of-concept three-dimensional reconstruction of the giant Mimivirus particle. Three-dimensional structure determination requires the assembly of many two-dimensional diffraction-patterns into an internally consistent three-dimensional Fourier volume. Since each particle is randomly oriented when exposed to the X-ray pulse, the relative orientations of the particles have to be retrieved from the diffraction data alone. We achieve this alignment using a modified version of the EMC algorithm [4]. For a new imaging method, validation is very important. We introduce two new validation methods inspired by X-ray crystallography and cryo-EM respectively. These methods confirm our results and provide a benchmark for future experiments.

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Ptypy - A python package for ptychography

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Ptychography, a scanning Coherent Diffractive Imaging (CDI) technique, has quickly gained momentum as a robust method to deliver quantitative images of extended specimens at diffraction-limited resolution. Following the pioneering work by Faulkner and Rodenburg [1], the algorithmic base [2-4] has grown rapidly to solve for various experimental limitations, from reduced signal [5,6], to inaccurate measurements [7,8], to various sources of diffraction data degradation, e.g. partial coherence effects [9], point-spread-function & air-scattering [10], and sample jitter [11]. While the essence of each algorithmic development is described in publications, exchange of implementation details, knowledge and expertise has remained limited between scientific groups.

This situation imposes a high entry-barrier for new groups and beamlines seeking to support state-of-the-art ptychography. In addition, validation of scientific results remains difficult if the reconstruction code is not distributed with the results.

Considering that many high-resolution nano-probe beamlines are planning to support ptychography as a standard, and that ptychographic algorithms have reached a sufficient level of maturity, it has become clear that the community needs broader access to state-of-the-art open-source reconstruction software. Here we present our own effort, *ptypy* (pronounced “tai-pai”), a python package developed in our group and aiming at offering implementations of all cutting-edge developments in the field. The capacities of our package will be demonstrated on widely diverse datasets, and the community will be invited to use it and to contribute to it.

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X-ray Scattering from Optically Trapped Nanoparticles

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A major barrier to applying coherent X-ray diffraction imaging to freestanding micro- and nano- scale objects is their tendency to freely move within the intense beam from synchrotron source. Typically such objects must be securely bonded to a substrate, which can alter their internal structures. While the forces moving such particles are not completely understood, we believe optical tweezers may be a solution. Optical tweezers provide a unique method to control small particles, ranging from several microns to tens of nanometers. Using optical techniques, these laser trapped particles can be manipulated and forces on the objects in the trap can be measured. Combined with phase modulation techniques, optical traps with different geometries and polarization can be generated, resulting in an accurate orientation of anisotropic particles [1,2]. We have built an apparatus of dynamic holographic optical tweezers which is compatible with x-ray diffraction imaging at beamline-34 of APS. In August we are going to test the x-ray diffraction from optically trapped micro- and nano- particles of both materials and biological sample origin with the goal of eliminating the barrier to studies of freestanding objects due to uncontrolled sample drifts. By observing the Bragg peaks, we can determine and optimize the stability of trapped particles. Then we will use coherent x-ray diffraction imaging [3] to image a freestanding sample explicitly selected by the user for the first time.

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Towards experimental 3D Coherent Diffraction Imaging from a large series of noisy diffraction patterns

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For 3D Coherent Diffractive Imaging e.g. of a macromolecule, a large series of coherent diffraction patterns from random sample orientations needs to be collected [1]. Even though corresponding analysis schemes have been devised [e.g. 2], an experimental demonstration of the method with realistically low signal and appropriate background level in diffraction space is still an open issue.

Here we report on a recent experiment at beamline 34-ID-C of the APS aiming at the collection of a large series of weak coherent diffraction patterns from varied orientations of a test particle (a cluster of 200-nm gold particles). The collected dataset contains 91×3000=273,000 diffraction patterns obtained for each of 91 rotation angles about a single rotation axis over a range of 90 degrees. The level of scattered signal in these patterns, which could be collected without a beam stop using the high-dynamic range Mixed Mode Pixel Array Detector (MMPAD) [3], lies within the range of expected signal levels from biological macromolecules at X-ray Free-Electron-Laser sources. We will report on the experiment and the initial analysis.

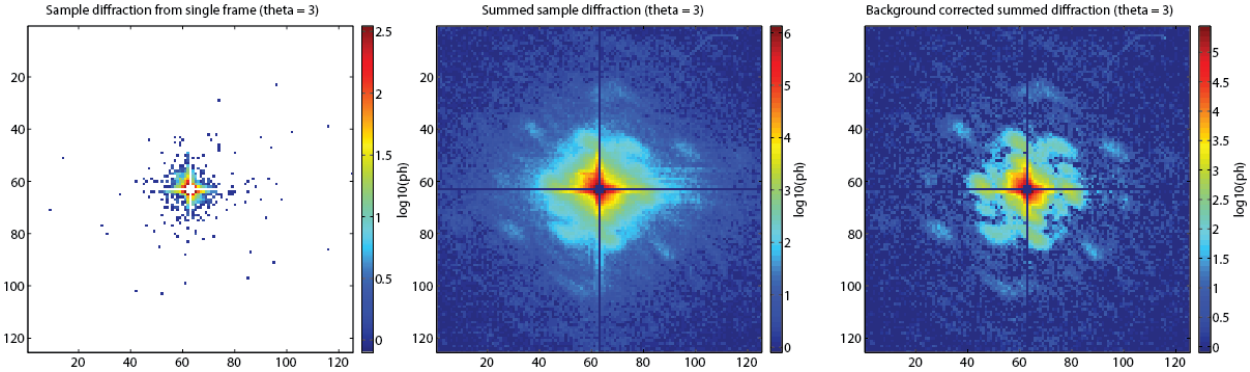

Figure 1: (Left) One out of 3000 collected diffraction patterns for a single projection angle. (Middle) Corresponding sum of 3000 patterns. (Right) Sum of 3000 patterns, with white-beam background (no sample) subtracted. Note that the colorscale for the left-most graph has been cropped to emphasize single-photon events. Direct beam contributions have been masked out.

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Soft X-ray Tomoholography: Exploration of Advanced Reference Structures

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In common mask-based Fourier transform holography (FTH) the reference wave is produced by a small pinhole, which is monolithically integrated with the sample [1]. Due to the high aspect ratio (1:10) of the pinhole it is not possible to record a tomographic dataset as the required rotation of the sample blocks the transmission through the pinhole already at small rotation angles. Recently, it was shown that a small gold sphere placed on an X-ray transparent membrane can be used to produce the reference wave [2]. This setup allows to rotate the sample and to collect a number of 2D projections for tomography.

We explore tomographic FTH with respect to use of more sophisticated reference structures. To this end, we apply coded aperture imaging (MURA) [3] and extended reference structures (HERALDO) [4] to tomoholography. This significantly increases the SNR of the 3D image reconstruction. Two demonstration experiments are presented where test structures are imaged with a lateral resolution of 65 nm.

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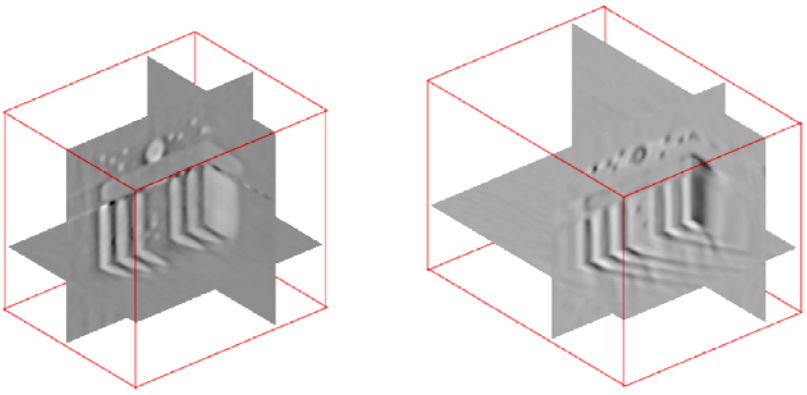

Figure 1: Three orthogonal slices of the tomogram of the test structure (Brandenburg Gate) imaged with the MURA (left) and HERALDO (right). The small dots above the Brandenburg Gate have a diameter of 80 nm.

Femtosecond speckle and coherence experiments at the CHG short-pulse facility DELTA

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The interaction of optical light pulses with electrons in an undulator can yield very short and coherent pulses of higher harmonic light (coherent harmonic generation □CHG concept). Using the CHG facility at the synchrotron DELTA we performed single pulse diffraction and speckle experiments with femtosecond long pulses of optical and UV light of wavelengths of 400 and 200 nm, respectively. The properties of the speckle pattern allow to deduce the coherence properties of the CHG radiation on a shot to shot basis. We find a high degree of spatial and temporal coherence of the CHG generated radiation when compared to the coherence properties of the spontaneous undulator radiation.

Single particle structure determination using cross correlation techniques

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We report on coherent X-ray diffraction experiments with the aim to determine the structure of a single particle when coherently illuminating a disordered ensemble of many particles. We discuss the influence of sample thickness and curvature of the Ewald sphere on the cross-correlation functions. The results for several model sample systems are discussed.

High-throughput imaging of cell organelles with an X-ray laser

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Structural heterogeneity interferes with crystallisation and causes systematic gaps in structural biology. Ultra-intense femtosecond pulses from X-ray lasers permit solving structures without crystals [1,2]. Every diffraction pattern is a unique structure measurement and high-throughput flash-diffractive imaging allows sampling the conformational space of heterogeneous structures. We demonstrate this in an experiment on carboxysomes [3,4]. Carboxysomes are heterogeneous, polyhedral cell organelles that faclilitate 40% of Earth's carbon fixation [5]. A new aerosol sample-injector allowed us to record 70,000 low-noise diffraction patterns in 12 minutes at the 120 Hz repetition rate of the LCLS. The diffraction data shows that the size distribution is preserved during injection. We computationally separate different structures directly from the diffraction data, automate phase retrieval, improve resolution, and avoid reconstruction artefacts. These advances lay foundations for accurate, high-throughput studies on structure and structural heterogeneity in biology and elsewhere.

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Toward XFEL imaging of Au-labeled icosahedral virus

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XFEL offers new opportunity of imaging bio-macromolecule by "diffraction before destruction". However, due to the photon flux available at XFEL sources, the diffraction patterns from biological macromolecules are still limited by shot noise. To overcome the challenge of recovering phases from noise-limited diffraction patterns, an Au-template method was proposed by one of us [1]. Based on such notion, we are purifying icosahedral virus particles of various sizes ranging 30 nm to 100 nm and labeling them with nanogold (Au) particles for XFEL experiments. Cryo-electron microscopy of the HBV virus like particle followed by 3D reconstruction to 7 Å have revealed nanogolds situated on 120 unique sites, ensuring the quality of the labeled virus. Simulations are being performed to study the resolution as a function of XFEL photon density or electron dose in the presence of Au vs the absence of Au, aimed for uncovering the role of Au nanoparticle in increasing the phase retrieval efficiency in 3D reconstruction from XFEL diffraction patterns.

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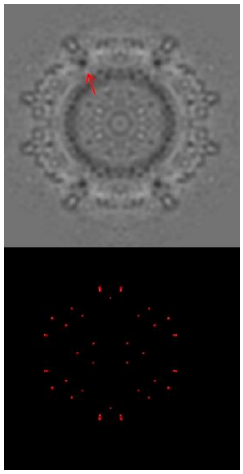

Figure 1: Cryo-EM of Au-labeled HBV virus like particle; in the upper, a 2D slice of the difference map between core HBV-Au VLP and core HBV VLP, and the lower one is a 3D difference map where the Au sites are colored in red.

Dynamical artifacts in Bragg coherent diffractive imaging

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The iterative phase retrieval algorithm used in Bragg coherent diffractive imaging (BCDI) technique is based on the assumption that x-rays diffract kinematically from a crystalline particle, in which case the diffracted far-field wavefield can be formulated into a Fourier transform of the shape and the strain field of the particle. For a relatively large particle size close to or above 1 μm, however, this assumption becomes invalid because dynamical diffraction effects have to be considered in such cases. Here we report a theoretical study on the reconstruction artifacts in BCDI introduced by dynamical diffraction effects, using synthetic data computed by a forward model published recently [1]. We show that, the phase, or the strain field in other words, is more sensitive to the dynamical diffraction effects. In the extreme case where dynamical diffraction is dominant, the iterative reconstruction algorithm fails; it does not converge to the true object function at all. We also discuss the conditions under which dynamical artifacts are negligible.

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Multiplexed coherent X-ray photon correlation spectroscopy measurements using micro-slit arrays fabricated by photolithography

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Abstract:
Increasing experimental throughput and *in-operando* characterization of different components of functional materials are among major challenges faced by the global synchrotron community. Aiming to address these challenges, we propose and demonstrate a multiplexing scheme based on micro-slit arrays fabricated by photolithography. This scheme, which in principle is applicable to all in-line synchrotron techniques, allows parallel measurements and enables better measurement statistics or probing heterogeneous structure, dynamics, or elemental composition simultaneously. As an example to illustrate this scheme, we utilize ultra-small angle scattering based X-ray photon correlation spectroscopy to study the equilibrium dynamics of a simple colloidal suspension. We found that the relaxation time follows a monotonic decay as scattering vector increases and the inverse of the effective diffusion coefficient exhibits a peak that mimics a static structure factor, consistent with previous findings. More importantly, the multiplexing scheme leads to a better measurement statistics that place the measurement uncertainty within instrumental resolution. We will also discuss the potential of the multiplexed scheme in elucidating the response of different components of a heterogeneous sample under *identical* experimental conditions in *simultaneous* measurements, which potentially could open a new paradigm for *in-operando* characterization of heterogeneous functional materials.

Development and Demonstration of visibility-based X-ray Photon Correlation Spectroscopy

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When coherent X-rays impinge upon a disordered system, a grainy scattering pattern called speckle pattern is observed. If the system evolves with time, the corresponding speckle pattern also changes. Temporal changes in the speckle patterns therefore provide information on the dynamics of system. This technique, which is called X-ray photon correlation spectroscopy (XPCS) [1,2], has shown the potential to access dynamic properties of various materials, such as colloidal suspensions, block copolymers, supercooled liquids, and antiferromagnetic materials.

Although XPCS is a powerful technique for materials science, it has a limitation of time resolution: dynamics faster than the frame rate of a detector cannot be measured. When a two-dimensional (2D) detector is used, the typical time resolution of XPCS is limited to the order of milliseconds.

For improving the time resolution of XPCS, we have extended speckle visibility spectroscopy (SVS) in the region of visible light [3] to the region of X-rays (X-ray SVS; XSVS) [4,5]. Since the minimum exposure time of the scattering patterns determines the time resolutions of XSVS and SVS, micro- or nano- second dynamics can be measured even with a 2D detector. Thus, XSVS has potential to bridge the time gap between XPCS and inelastic neutron/X-ray scattering techniques, and will be one of the promising tools for various fields in science expected with the next generation synchrotron X-ray sources, such as diffraction limited storage rings and energy recovery linac based X-ray sources.

In this presentation, we will describe the principle of XSVS and show the result of the application of XSVS to Brownian colloidal suspensions.

This study was performed under the approval of JASRI (2011A1112, 2011B1131). We acknowledge Drs. N. Yagi and N. Ohta for their kind support in performing experiments.

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Counting dislocations in micro-crystals with coherent x-rays: ex-situ and in-situ studies of the plastic deformation of InSb micro-pillars

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Coherent x-ray micro-diffraction was used to detect and count phase defects (stacking faults, SFs, left in the crystal after the glide of partial dislocations) introduced by mechanical deformation of InSb single-crystalline micro-pillars (Fig.1a). Diffraction patterns were recorded by scanning the coherent micro-beam along the pillars axis: peak splitting is observed in the diffraction pattern associated to the top region, in agreement with the presence of a few SFs located in the upper part of the deformed pillars (Fig. 1b). Simulations of coherent diffraction patterns were also performed considering SFs randomly distributed in the illuminated volume: they show that not only the number of defects but also the size of the defected volume influences the maximum intensity of the pattern, allowing for a good estimation of defects number and defected volume size [1]. These results are promising for the study of semiconducting devices in which defects can affect the performances.

Similar measurements were performed in-situ, during mechanical compression, to detect the first lattice defects, i.e. the first events of the plastic deformation appearing in InSb micro-pillars.

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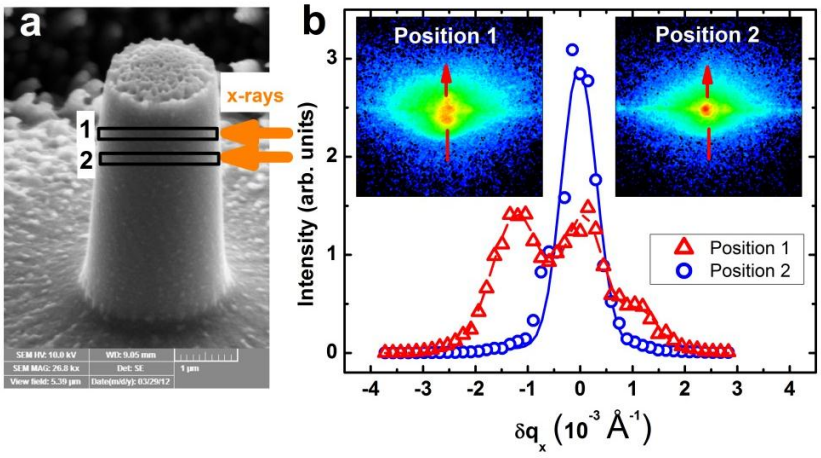

Figure. 1: a) Scanning Electron Microscopy image of the studied pillar. The volumes probed at positions 1 and 2 are indicated. b) Insets: Coherent diffraction patterns on the 202 Bragg reflection recorded at positions 1 and 2. Graph: Projection of the patterns along vertical direction of the detector.

XMDYN: Modeling radiation damage of XFEL irradiated samples

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High-resolution x-ray imaging of nanosize biological samples is one of the most important goals of the research with X-ray free electron lasers (XFEL) [1]. Outstanding results have already been achieved in serial nano-crystallography [2]. There has been also a significant progress in the field of single particle imaging [3]. However, radiation damage is still a limiting factor, in particular for non-periodic objects. This requires thorough theoretical investigations of the time evolution of the irradiated samples.

Here we report on XMDYN [4], our molecular-dynamics based tool to model the dynamics of finite samples irradiated by high intensity x-ray pulses. First we describe the theoretical approach used. To validate the model we then show predictions of the model as compared to experimental results. New developments such as the on-the-fly connection to the atomic physics XATOM toolkit [5] enabling accurate treatment of heavy elements and a possible code extension towards large-scale calculation are also discussed.

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Imaging of Live Cell in Micro-Liquid Enclosure Array by Single-Shot X-ray Free Electron Laser Diffraction

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In imaging of biological sample, it is essential to keep the sample close to natural state, such as in solution. By freezing the sample in time using the femtosecond pulse duration, X-ray free electron laser (XFEL) can overcome the problems of molecular vibration and radiation damage which limit achievable resolution. Imaging technique using XFEL thus has great potential for high-resolution imaging of biological samples.

In this study, we present a method for capturing snapshots of live cells kept in a micro-liquid enclosure array (MLEA) by single-shot XFEL diffraction [1]. MLEA is shown in Figure 1 (a). MLEA consists of two custom-made silicon microchips. Living bacteria are placed between 100-nm-thick silicon nitride membranes. To reduce parasitic scattering and prevent the sample from overlapping, thickness of the solution layer is controlled by SiO_2 spacer.

We performed live cell imaging experiment using the SPring-8 Angstrom Compact Free-Electron Laser (SACLA). We exposed each enclosure to a single X-ray laser pulse from the SACLA and recorded a coherent diffraction pattern from enclosed bacteria with a clear fringe extending up to a 28-nm full-period resolution. The reconstructed image (Figure 1 (b)) reveals living whole-cell structures without any staining, which helps advance understanding of intracellular phenomena.

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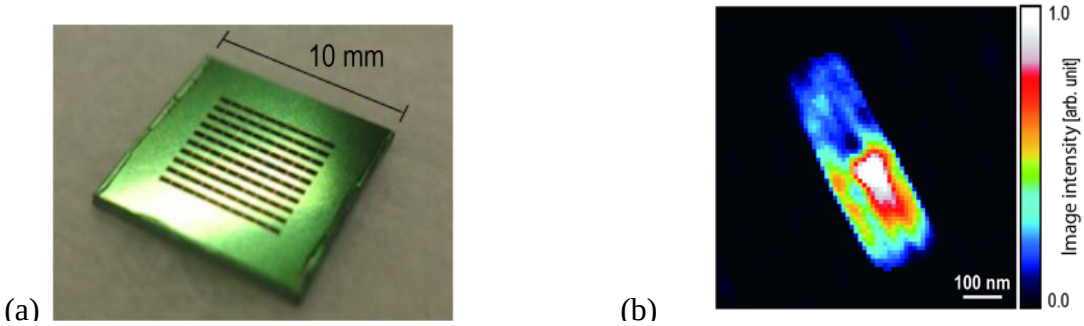

Figure 1 (a) Photograph of MLEA chip with 100 enclosures.
(b) Reconstructed image of enclosed bacteria.

Forbidden Reflections in X-ray Crystal Truncation Rods: Using Surface Reference Waves to Distinguish Charge and Vibrational Asymmetry in Silicon

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X-ray reflections from diamond crystal structures with Miller indices that satisfy h+k+l = 4n+2, where n is an integer, are considered to be forbidden by crystal symmetry. However, the asymmetry of the valence charge distribution as well as anharmonic vibrations break the symmetry and result in weak, but non-zero intensities for these "forbidden reflections". Due to the absence of phase information, considerable effort, involving combined x-ray and neutron scattering studies as well as temperature-dependent measurements, was previously required to determine the relative contribution of charge and vibrations to these reflections. In the present work, we demonstrate that useful phase information can be gained in x-ray reflectivity and crystal truncation rod measurements where there is interference between waves scattered from the bulk and the surface. In this manner, surface reference waves can be used to determine the charge and vibrational asymmetry in the bulk crystal. These effects are demonstrated with synchrotron x-ray scattering measurements performed in ultra-high vacuum on Si(111)7x7 surfaces with and without Ag films. Understanding the properties of the forbidden reflections in diamond crystal structures not only provides greater insight into the crystal bonding and vibrations, but can also lead to better models for surface structures determined by x-ray scattering.

Support from the National Science Foundation under grants DMR-0706278 and DGE-1069091 is gratefully acknowledged. The Advanced Photon Source Sector 6 beam-line at Argonne National Laboratory is supported by the US-DOE under Contract No. W-31-109-Eng-38.

Optimization of X-ray Ptychography using the Excalibur detector at I-13 coherence beam line at Diamond

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Ptychography is an imaging technique which is fundamentally a type of coherent diffractive imaging [1,2]. The sample is scanned in two dimensions by a micro-probe collecting diffraction patterns at each point, after which the image is reconstructed by employing iterative algorithms. We will document our first successful steps in optimizing the technique with the new Excalibur detector employed at I-13 coherence beamline at the Diamond Light Source [3]. Simultaneously we carried reconstructions of diatoms in order to explore the scope and potential of ptychography. We aim to achieve faster scan rates thus inch towards carrying ptycho-tomography at the facility. Future work is aimed at the induction of an updated version of Excalibur detector, which will feature “back to back read-out” and “color mode”, opening exciting avenues like multi-wavelength imaging and ultra-fast on the fly scanning ptychography . Hard X-ray ptycho-tomography, at 10 keV and above, will open new avenues for the investigation of denser and thicker samples than has been possible up to now.

We acknowledge financial support through the European Research Council (ERC, starting grant “OptImaX”).

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(a) (b)

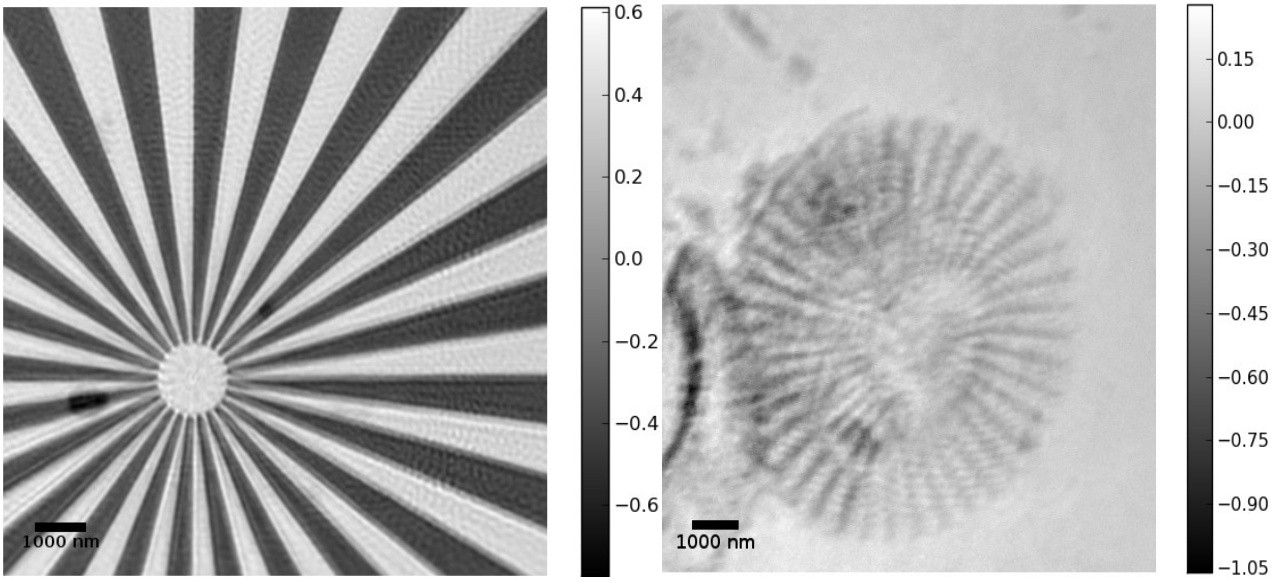

Fig1. (a) A Siemens star test sample, demonstrating better than 100 nm resolution. (b) A diatom fragment. Both phase images were obtained at the I-13 beam-line with a photon energy of 9.7keV and using the Excalibur detector.

Anomalous dynamics prevent the glass transition in a hard-sphere colloidal suspension

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We describe results obtained by multi-speckle X-Ray Photon Correlation Spectroscopy on colloidal suspensions of Hard Spheres (HS) upon approaching the glassy state. HS are widely used as a model system to study how the fundamental structural relaxation (α-relaxation) freezes out as the glass transition is approached from the supercooled state. Our results show that the HS system is not necessarily ending up in a true non-ergodic state, even at concentrations well above the commonly accepted glass transition concentration Φ_C found in other experiments and predicted by Mode Coupling Theory (MCT). In fact, we observe an aging, anomalous dynamics that is dominated by avalanches with intermittent periods of arrest and pronounced dynamical heterogeneity [1]. The departure from MCT happens already in the supercooled state below Φ_C and we ascribe the behavior to stresses that build up in the sample and relax the structure. Recently, other studies on colloidal systems have demonstrated an unexpected high sensitivity of the relaxation dynamics on details of the sample preparation, for instance concerning the application of shear [2,3]. We conclude that the observed dynamics in the HS system at high concentration is due to a competition between stress relaxations and an arrested glassy state.

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X-ray Correlation Spectroscopy from dewetting polymer bilayer films

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The initial microscopic mechanism of dewetting of Poly(4-bromo styrene) thin film deposited on an immiscible and non wettable Polystyrene sublayer on Silicon substrates was investigated using diffuse X-ray scattering at grazing incidence in conjunction with two-time X-ray Correlation Spectroscopy (XPCS). Selective excitation of the top and bottom interfaces using an x-ray standing wave similar to that reported in [1,2] allowed the dewetting process to be measured as a function of depth. The early stages of dewetting were monitored by measuring the surface diffuse scattering from the top and buried interfaces in the polymer/polymer bilayer. Different modes of thickness fluctuations in the bilayer films were observed for the first time at the onset of dewetting measured as function of film thickness and viscosity. X-ray results are also compared to atomic force microscopy images of the final dewetted patterns.

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Higher Order X-ray Intensity Correlations in Amorphous Materials

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Despite intensive studies in the past decades, the local structure and order of disordered matter such as liquids and glasses remains widely unsolved. Simulation studies proposed that bond orientational ordering defines various properties of the material and may play a key role e.g. in crystallization of hard spheres systems [1]. In contrast to classical nucleation theory, an increased orientational order was observed before critical nuclei start to grow, but experimental proof is rare.

X-ray Cross Correlation Analysis (XCCA) was demonstrated recently to be a promising tool to reveal the sample's local orientational order in liquids and glasses [2]. By studying angular correlations in coherent diffraction patterns a dominant orientational order can be determined [3].

Here we want to show the potentials of XCCA for studying the local order in amorphous sample systems. In a first step, we apply XCCA to simulations of model structures and define experimental limitations [3]. Afterwards experimental results from two-dimensional colloidal model systems are discussed [4]. Special attention is paid to the investigation of the change of local order in colloidal hard-sphere systems at different volume fractions ranging from the liquid to the glassy state. In the "supercooled" state an increase of order is observed that vanishes for amorphous systems. This increasing order is furthermore accompanied by a slowing down of the particle dynamics. Finally, we discuss the potentials of XCCA to reveal transient structures from ultrafast x-ray scattering at Free-Electron Laser facilities and show first results from liquid and charge stabilized sample systems.

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Inverse Problem Theory for X-Ray Diffraction of Nano-particles

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In this work, we present the theory and accompanying algorithm for the recovery information pertaining to the positions of the atoms in nano-particles, or arrangements of nano-particles, from X-ray diffraction intensity data. The theory is based on the linearization of the intensity functional followed by the application of a proper diagonal pre-conditioner and a linear filter, leading to a well-conditioned problem. In particular, we provide guarantees as per when it is possible to proceed to the inversion of the data and reach a unique solution for the location of the atoms. The importance of such information is crucial as shown in Figure 1 where two different distributions of atoms give rise to roughly identical intensity pattern, implying that it is impossible of their locations from such information.

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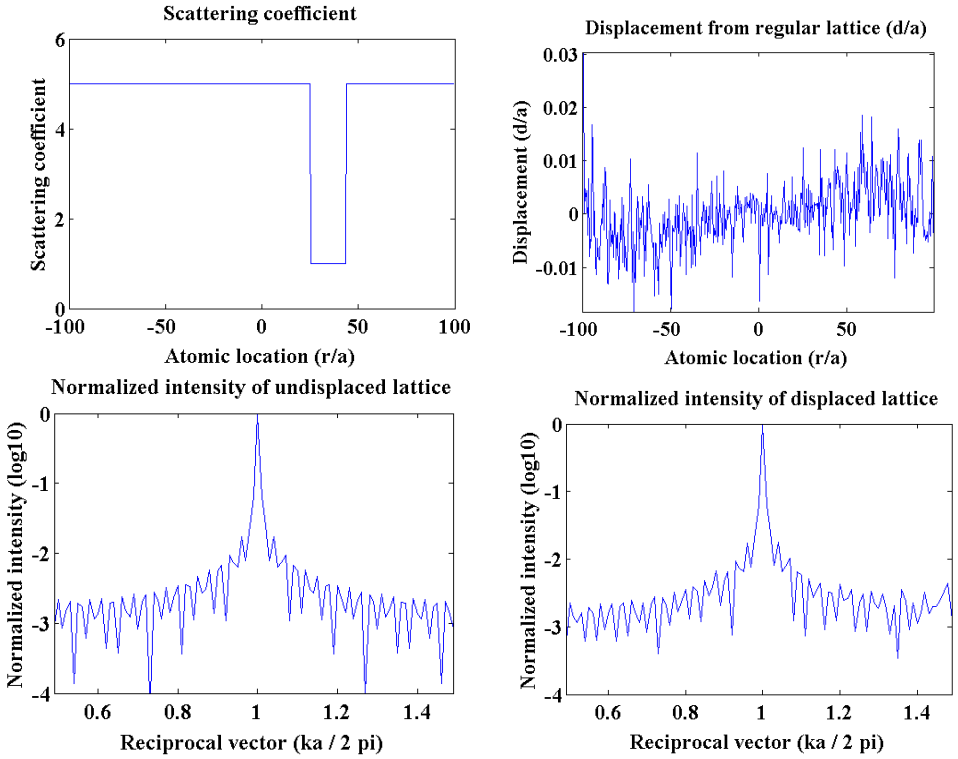

Figure 1: Example of an ill-conditioned inverse problem (noiseless); Bottom left: normalized intensity ($\log_{10}$) around the first Bragg peak from a cluster of 400 scatterers with scattering coefficients shown on the top-left figure, and located on a regular grid. Bottom right: normalized intensity ($\log_{10}$) around the first Bragg peak from the same cluster for which the location has been perturbed by an amount shown in the top-right figure. Both pattern appear almost indistinguishable despite the fact that they arise from significantly different distributions of atoms (up to 3% of the lattice constant displacement at each location).

Measuring Flows and Velocity Gradients with XPCS

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Conventional coherent Xray Photon Correlation Spectroscopy (XPCS) does not allow for the measurement of uniform flows. By adding a static randomly distributed sample as a reference, such uniform flow flows can be measured. This heterodyne technique is used to measure the flow across a channel, and probe its boundary conditions.

This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357.

Simulation of Paraxial Wave Propagation for Zone Plate Optics

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Simulation of wave propagation through axially symmetric system can be done by Hankel transform (HT) in cylindrical coordinates. Quasi Discrete Hankel Transform (QDHT) algorithm has been developed to perform HT [1-2]. Axial value can be obtained by sampling theorem [3]. A rapid HT based simulation tool is developed for paraxial wave propagation and properly used for near and far field propagation in order to avoid aliasing caused by sampling. The focal spot profile with high resolution down to 1 nm can be rapidly simulated. Stacking zone plates [4-6] up to four in further proximity is simulated. Various schemes of zone plates [7-10] for high efficiency and resolution are also simulated and compared.

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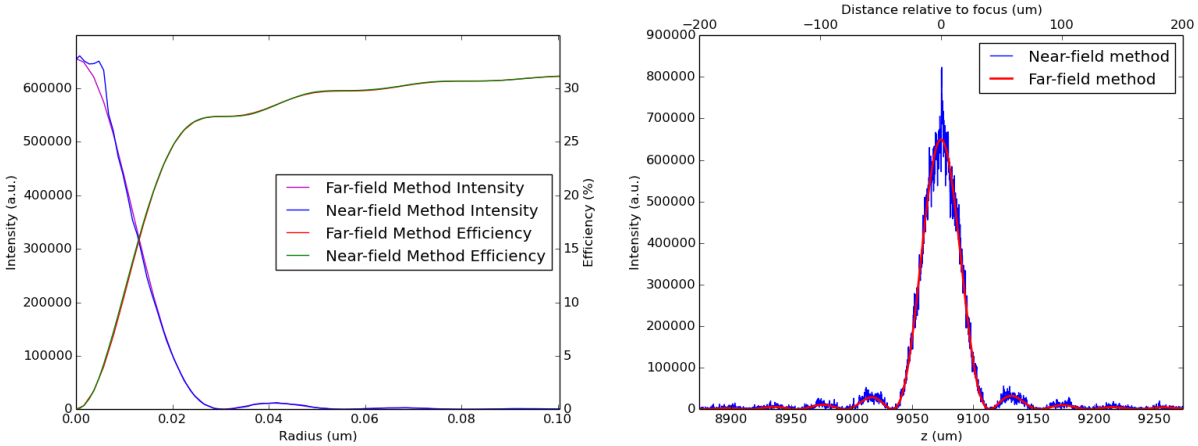

Figure 1: Comparison of two methods for (left) radial intensity distribution and energy efficiency near the focus and (right) depth of focus of a D = 45 μm, Δr = 25 nm zone plate.

Method to enhance the resolution of x-ray coherent diffraction imaging for non-crystalline bio-samples

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To circumvent the problem of radiation damage when using an x-ray coherent diffraction imaging experiment to resolve the structure of biological samples, we propose a method to add objects made of heavy atoms with the bio-samples or we load the samples on a template made of heavy atoms. This template method is shown by a numerical simulation (including shot noise) to be able to resolve the structure of a virus better than without the template [1]. A counter-intuitive result is obtained, where heavier templates have a better resolution, even if the diffraction intensity of the bio-sample is much smaller than the noise intensity. In addition, the method also helps to greatly increase the efficiency of phase retrieval. We also provide a way to estimate the error to be expected if a particular experimental setting were chosen once the charge ratio between the sample and the template is estimated. Hence, this method will also help experiments choose the optimal setting for the best resolution with minimal radiation damage.

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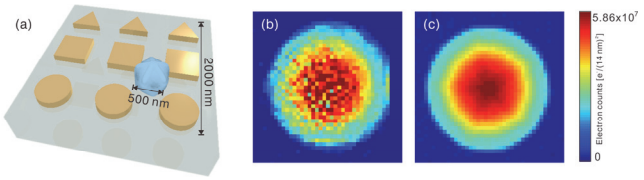

Figure 1: (a) A Mimivirus is put on a template made of gold. (b) The reconstructed projection of an isolated Mimivirus from noisy diffraction intensity. (c) The reconstructed projection of a Mimivirus measured with the template from noisy diffraction intensity.

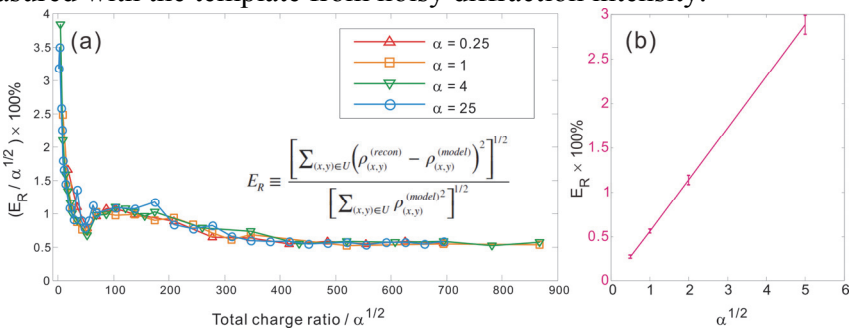

Figure 2: (a) Normalized E_R versus total charge ratio under different exposure time shortening factor α, where E_R, definition shown within the panel, indicates the quality of the reconstruction. (b)The average and corresponding error bars of converged E_R as the total charge ratio goes to infinity versus the square root of α.

Signal amplification in coherent diffractive imaging using diffractive optics

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We investigate the use of diffractive optics for signal amplification in coherent diffractive imaging (CDI). In particular, a specific geometry is analyzed where a Fresnel zone plate is placed up-stream from the sample. The experimental geometry is similar to Fourier transform holography (Fig. 1). The interference of the two beams increases the high spatial frequency signal detected on the CCD in comparison to plane wave CDI. The data is obtained using an extreme ultraviolet laser with a high degree of coherence that makes it well suited for coherent imaging [1,2]. There are several challenges associated with uncertainty in the reference wave and field of view tradeoffs. Despite these challenges, the technique shows the potential to increase high-spatial frequency signal and alleviate problems associated with the limited dynamic range of current CCDs effectively altering the signal falloff rate associated with these types of scattering experiments.

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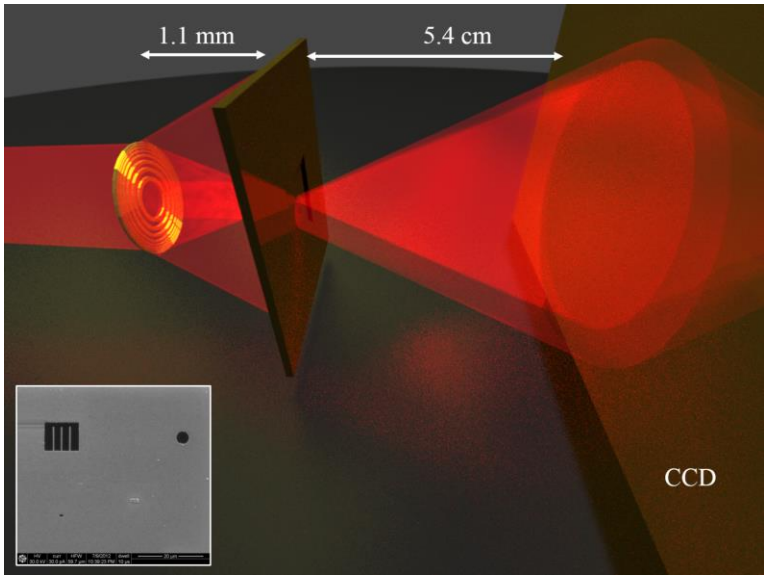

Figure 1: Illustration of the proposed CDI experiment and SEM image of the experimental sample (inset).

Ultrafast diffractive imaging with coherent electron pulses: light-induced ordering phenomena of functionalized nanoparticles

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We report a femtosecond electron diffractive imaging technique that allows to characterize a 2D film of homoligand self-assembled gold nanoparticles with sub-nm spatial resolution and sensitivity to the carbon and hydrogen atoms in the ligands. Experiments are carried out using a high-flux UED set-up capable of performing coherent small-angle fs-electron diffraction [1]. Control on the dynamics of the nanoparticles and the arrangement of the ligands is achieved via ultrafast laser excitation, demonstrating that ordering phenomena can be triggered by light in these systems.

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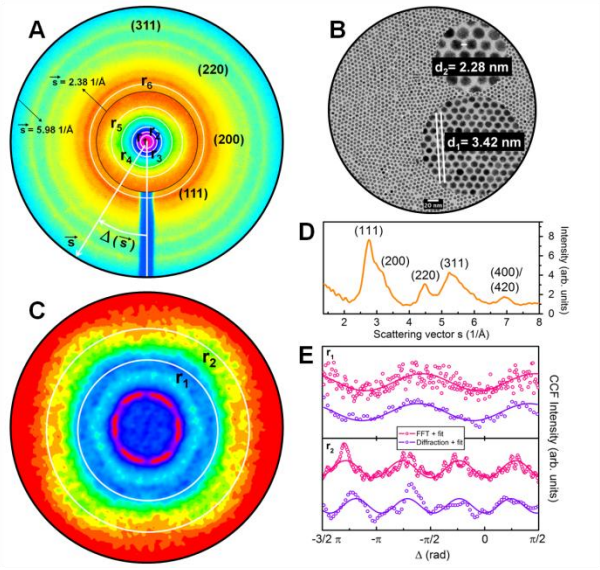

Figure 1. **a**) Electron speckle diffraction pattern of homoligand gold nanoparticles. **b**) TEM image of the homoligand gold nanoparticles 2D-assembly. **c**) 2D-FT of the TEM image displayed in (**b**). **d**) The diffraction from the gold centers at large-angle is integrated over 2π and plotted as a function of the scattering vector. **e**) Angular intensity modulations of the speckle pattern from the rings $\mathbf{r_1}$ and $\mathbf{r_2}$ indicated respectively in (**a**) (violet profile) and (**c**) (pink profile). The modulations are a fingerprint of local symmetries in the polycrystalline nanostructured sample.

Single phase grating interferometers for probing x-ray beam coherence area

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Single phase-grating interferometers were developed for probing transverse (spatial) coherence of the x-ray beam. At first, a 2-D checkerboard phase-grating interferometer [1] for measuring beam coherence along multiple (four) directions is described. Visibilities of interferograms measured at Talbot self-imaging positions downstream of the grating were used to obtain the complex coherence factor (CCF) and the coherence lengths not only along the vertical and horizontal directions but also along the 45° and 135° directions from the horizontal direction. Recently, we have also developed a circular grating interferometer [2] to measure the coherence lengths along all transverse directions simultaneously and therefore to map the transverse coherence area of the x-ray beam. The transverse coherence lengths obtained by both interferometers were verified by the wavefront simulation and analytical calculation. Application of this technique for transverse coherence characterization of x-ray sources is important for selecting appropriate sample size and sample orientation as well as for identifying and overcoming the partial coherence effects in an experiment.

Use of the Advanced Photon Source and Center for Nanoscale Materials, Office of Science User Facilities operated for the U.S. Department of Energy (DOE) Office of Science by Argonne National Laboratory, was supported by the U.S. DOE under Contract No. DE-AC02-06CH11357. We acknowledge Dr. Ralu Divan for her assistance during the grating fabrication.

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Correlated X-ray Scattering: How to Extract High-resolution Information from Solution Measurements

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Correlated x-ray scattering (CXS) is an emerging technique, which has the potential to recover single particle information from solution measurements. In a solution, particles are randomly oriented, however if photons scatter into multiple pixels from a single particle in solution, then these photons are correlated. By averaging the correlations from many exposures of the solution, one can extract a correlation function related to the structure of a single particle from a background noise of uncorrelated photons. This is done by calculating angular correlations on the detector. It is critical that the x-ray exposure is short relative to the rotational diffusion time.

We have conducted CXS experiments at xFEL facilities and a micro-focus synchrotron beamline on silver and gold nanoparticles (NPs) in solution. Indeed we have been able to recover the single particle correlation function for the individual NP even though each exposure was done on a solution of many randomly oriented NPs. With standard powder diffraction one can easily determine that the structure of each NP is face-centered-cubic, however CXS reveals higher order information about the structure and symmetry of the NPs, such as the possibility of stacking faults, and multi-twinning within the crystallites.

In comparison to standard SAXS/WAXS and powder diffraction measurements, the parameter space for the CXS function is huge and can provide additional atomic constraints for models. The ability to precisely and accurately recover CXS from solution measurements will make possible the refinement of atomic models of proteins and biomolecules which can only be observed in solution phase due to limitations on crystal synthesis and imaging technology.

Coherent Diffractive Imaging of FePt Nanocrystals Embedded in a Matrix

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Coherent Diffractive Imaging (CDI) is a method that has been extensively developed by our group over the past 10 years [1,2] to investigate the structure and strain within an individual nanocrystal. CDI has been successful for nanocrystals in the 200 nm size range, but until recently failed for the 20 nm size-range, accessible by chemical synthesis. This CDI investigation of 17 nm FePt nanocrystals demonstrates that we can now overcome such limits through the use of a novel stabilisation method, developed within. Furthering the knowledge and understanding of the fundamental properties of FePt nanocrystals, in their more industrialably favourable face-centered tetragonal phase, will enable key applications in medicine [3], catalysis [4] and information technologies. The CDI investigations were carried out at the Advanced Photon Source 34-ID-C beamline, through imaging diffraction patterns of an isolated FePt nanocrystal embedded within a matrix. The nanocrystals were embedded in the SiO_2 matrix via a 'direct drop' method, developed within our group, and annealed at temperatures up to 800 °C to create the matrix and to phase transform the nanocrystals (from the as-sysnthesised face centered cubic phase to the face centered tetragonal). This amorphous matrix not only stabilises the nancrystals, preventing any rotational movement on exposure to the beam, it also acts as a protective coating preventing the nanocrystals from aggregating/oxidising during exposure to the high temperature of the annealing phase. CDI scans of an individual FePt nanocrystal were taken on the rocking curve of the selected face-centered tetragonal (111) Bragg peak and multiple Bragg reflections were measured, accumulating a 3D diffraction data, an example of these peaks from a frame of the rocking curve is shown in Fig. 1. The full data set was phased and the diffraction pattern was inverted, into a 3D image of the crystal. This technique is particularly sensitive to any strains present in the crystal and used to image complex density maps.

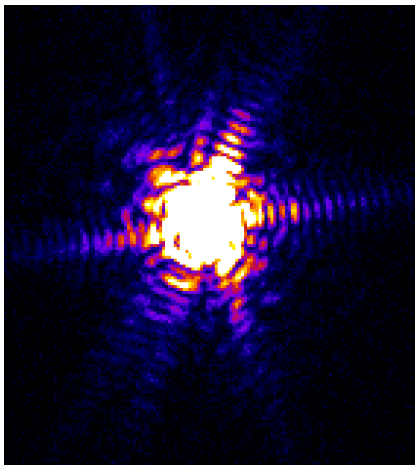

Fig.1.: Bragg diffraction frame of an isolated FePt nanocrystal

We would like to thank the beamline scientists (including Ross Harder) at the APS, 34-ID-C beamline, for their input with this investigation.

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PtychoLib: Parallel Ptychographic Reconstruction

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Recent advances in microscopy, in terms of higher spatial resolution and faster data acquisition, require increasing efficiency and accuracy of specimen reconstruction methods. Ptychography is a phase retrieval technique that is able to reconstruct the image of a specimen from a sequence of diffraction patterns, without the need for lenses in visible light, X-ray, and electron microscopes. Despite the potential for ptychography to deliver high-resolution images, there is a lack of freely available software for data management and online analysis to perform image reconstruction while the data is being acquired. Waiting to post-process datasets offline results in missed opportunities. PtychoLib is a parallel library for real-time ptychographic phase retrieval. It was able to achieve more than 200x speedup over an existing implementation of the extended ptychographical engine (ePIE) method. The library uses a hybrid parallel strategy to divide the computation between multiple graphics processing units (GPUs), then employs a novel technique to merge the result into one coherent phase contrast image. Results are shown on a simulated specimen, and a real dataset from an experiment conducted at the Advanced Photon Source.

Current and future applications of state-of-the-art photon counting detectors at I13

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The I13 beamline at Diamond Light Source opens exciting new opportunities for direct and reciprocal space imaging in the hard X-ray (6-25keV) energy range. Whilst in direct space, scintillator based detection is suitable, the high dynamic range needed for high resolution coherent diffraction techniques necessitates photon counting detection. EXCALIBUR [1] (STFC, Diamond Light Source, CERN) is a high resolution (55 micron pixels), 3 megapixel (11.4 x 9.9 cm active area) photon counting detector based on the MediPix 3 chip series with a readout system designed and implemented by the Diamond-STFC collaboration. The 1MHz photon counting rate combined with the 1kHz frame aquisition rate, makes EXCALIBUR perfect for the range of high coherence experiments possible at I13 including: CXDI[2], holography and XPCS[3]. This presentation will show results from the first experiments using this detector for high resolution ptychography of both test targets and real samples, and show commissioning test results for the time resolution.

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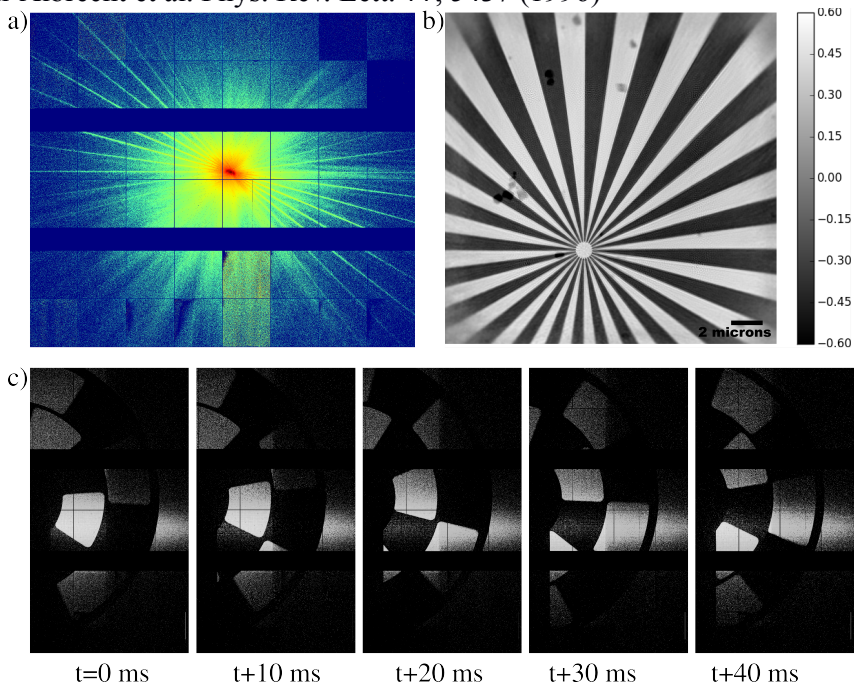

Figure 1: a) A log intensity plot of the sum of 10 recorded ptychographic data points from a Siemens star sample filling the full acceptance aperture.b) The phase of the reconstructed object transmission function from the same data set as a). c) A 100Hz time series collected in burst mode whilst an optical chopper rotates between the beam and the detector demonstrating EXCALIBURs' high time resolution.

Characterising Structural disorder in bionanocrystals at the Synchrotron and X-ray Free Electron Laser

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Understanding damage, disorder and crystalline defects is a topic of critical importance for structure retrieval in protein crystallography and optimization of crystal growth. Utilizing Bragg coherent diffractive imaging at the Advance Photon Source (APS), we have begun to investigate the strain distribution in the crystalline lattice of biologically relevant crystals with a view to gaining a better understanding of the factors affecting crystal growth. As well as applications in, for example, the development of new treatments for malaria, controlling the size and quality of biological crystals is also of direct relevance for the pharmaceutical industry. Another recent major development in protein crystallography has been the use of XFELs for structure determination from nanocrystals. Here we present XFEL diffraction data collected from nanocrystals of β-hematin, the synthetic form of hemozoin which is a by-product of the malaria parasite. Through structural analysis of femtosecond nanocrystallography data we observe significant deviations from the 'standard' textbook structure for this molecule which is based on synchrotron diffraction data. Our findings have implications for the interpretation of XFEL data from nanocrystals and suggest that, for some samples, we need to develop fundamentally new approaches to structure retrieval at XFEL's.

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Statistical hit finding for XFEL biosamples

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The investigation of 3D structures of biomolecular complexes at XFEL facilities results in the need to handle a significant amount of 2D diffraction patterns. In many cases most of the recorded patterns are representative of background events rather than actual sample. When the experimental conditions are pushed to the limits, distinguishing between the two for individual patterns might prove a challenge.
We have developed a general method in hit finding, assuming a static background model which is only scaled by pulse intensity. Using an assumption of a Poisson distribution, the Fisher method [1] for meta-analysis is used, where each pixel constitutes a separate hypothesis (the null one being background). The particular case of experimental data from weak scatterers (RNA polymerase II, Fig.1) recorded by the small-angle detector [2] has been analysed. Progress in this area can demonstrate the feasibility of performing X-ray diffraction on single molecules. With a proper background model, averaging over multiple weak patterns for 3D assembly will eventually become possible.

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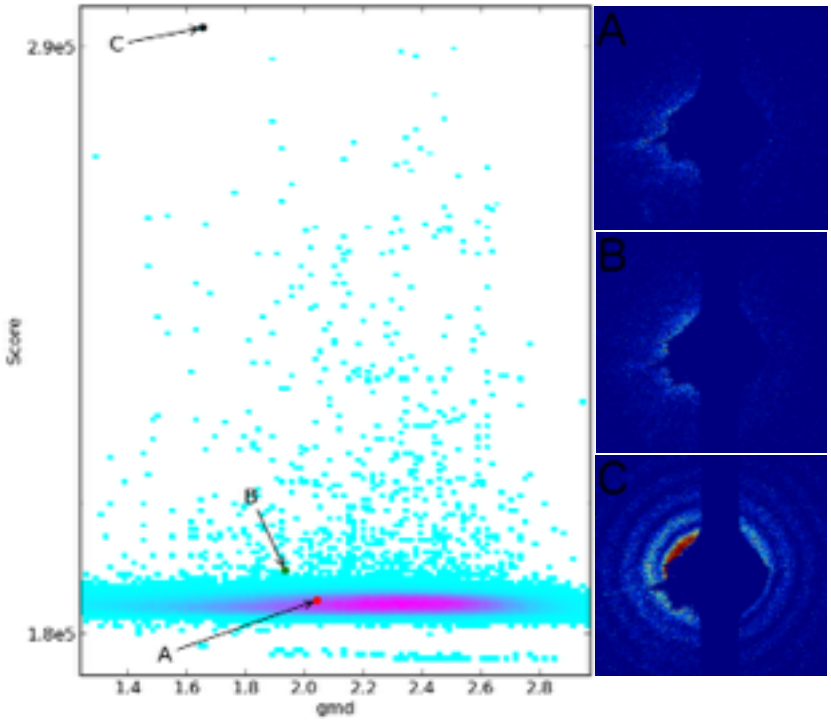

Figure 1: To the left: a density plot (log scale along y-axis and colour scale ranging from light blue to light purple) of the distribution of hit scores (using Fisher's method) versus pulse XFEL intensity as reported by the gas monitoring detector. Labels A,B and C refer to three specific diffraction patterns, also shown to the right. These are background (A), a weak hit (B) and a strong cluster hit (C). The colour scale used for the diffraction patterns is linear and goes from dark blu (0 photons) to red (photons ≥ 6).

Statistical description of four-point angular correlations: the concept of dominant orientational modes

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Despite remarkable progress in theoretical and experimental studies, the formation of thermodynamically driven microscopically highly inhomogeneous states (HIS) still belongs to the most intriguing scientific mysteries. In recent decades, the concept of the essential role of many-point correlation effects on stabilization of the HIS has emerged as a paradigm in understanding this problem. An important particular case of this spatial inhomogeneity is local bond orientational ordering. Modern experimental techniques on the basis of coherent scattering data, such as X-ray cross-correlation analysis (XCCA) [1] allow the direct determination of angular correlations in molecular disordered systems. Here we demonstrate the prospects of XCCA for the understanding of the importance of HIS using a statistical-thermodynamic approach. We argue that the main parameter for a statistical description of HIS is an ensemble averaged power spectrum of the cross-correlation function, which demonstrates a set of dominant modes. Calculations are performed both analytically and using molecular dynamics (MD) simulations for model systems with Dzugutov-type interaction adjusted for the creation of glassy-type quasi-equilibrium states [2]. XCCA applied to the simulated coherent scattering patterns from the MD samples show dominant modes responsible for the formation of non-commensurate structures, as found in glasses and quasicrystals. Strikingly, these modes exhibit a pronounced temperature-dependence indicating a critical-type behavior in the vicinity of the glassy-type transition.

This work has been supported by the excellence cluster 'The Hamburg Centre for Ultrafast Imaging – Structure, Dynamics and Control of Matter at the Atomic Scale' of the Deutsche Forschungsgemeinschaft.

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Simultaneous phase retrieval of object and probe for extended wavefronts

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Propagation x-ray imaging (at large defocus distances also denoted holographic inline x-ray imaging) requires either an aberration free or perfectly known probe. Since neither is the case, one generally falls back to dividing the intensities of the measured hologram by those of the recorded flat field. This raw data correction – at best only approximately correct [1] – is not capable of eliminating all artifacts resulting form an imperfect illumination, compromises quantitative contrast values and limits resolution (see Fig. 1).
Ptychography, originally invented for far field imaging techniques, allows for a simultaneous reconstruction of object and probe and hence overcomes the described difficulties. In contrast, for near field imaging this approach requires a deliberately distorted probe to provide convincing results [2]. To generate the necessary diversity for phase retrieval ptychography uses lateral shifts of an object through the X-ray beam. For full field holographic imaging this (lateral) diversity is insufficient for probes with moderate aberrations. However by adding defocus translations of the object and generalizing in particular the separability constraint of ptychography [3], we show that the diversity becomes sufficient to enable simultaneous reconstruction of object and probe also for extended wavefronts. This is demonstrated for parallel beam propagation imaging (ID19, ESRF) as well as nano-focused cone beam holographic imaging (P10, PETRA III, DESY). In summary, we can overcome the ubiquitous flat field correction and the associated artifacts (also demonstrated in this presentation) and put ptychography in a broader framework.

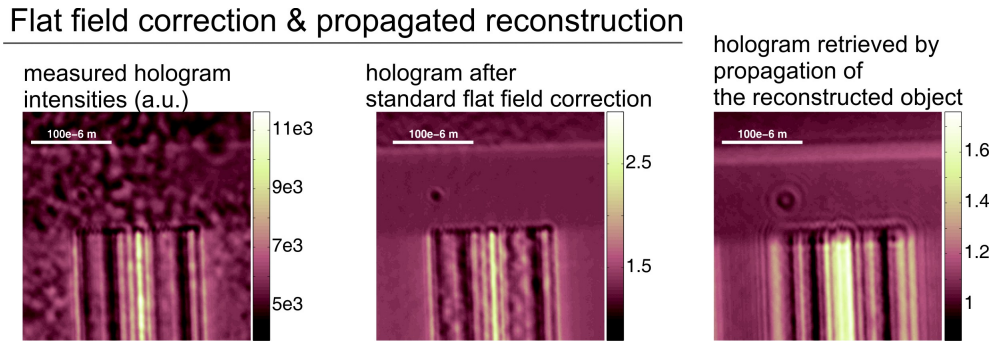

Figure 1: Left part: measured hologram, aberrations due to a non perfect illumination. Middle part: standard flat field correction, i.e. hologram divided by flat field. Right part: hologram produced by propagation of the reconstructed object.

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Three-Dimensional Coherent X-ray Diffraction Imaging of Whole, Frozen-Hydrated Cells

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A structural understanding of whole cells in three dimensions at high spatial resolution remains a significant challenge and, in the case of X-rays, has been limited by radiation damage. By alleviating this limitation, cryogenic coherent diffraction imaging (cryo-CDI) could bridge the important resolution gap between optical and electron microscopy in bio-imaging. Here, we report for the first time 3D cryo-CDI of whole, frozen-hydrated cells - in this case a *Neospora caninum* tachyzoite - using 8 keV X-rays. Our 3D reconstruction reveals the surface and internal morphology of the cell, including its complex, polarized sub-cellular architecture with a 3D resolution of ~75-100 nm, which is limited by the current implementation of our instrument. With the improvement of the instrumentation and the appearance of brighter X-ray sources worldwide, our work forecasts the possibility of routine 3D imaging of frozen-hydrated cells with spatial resolutions in the tens of nanometers.

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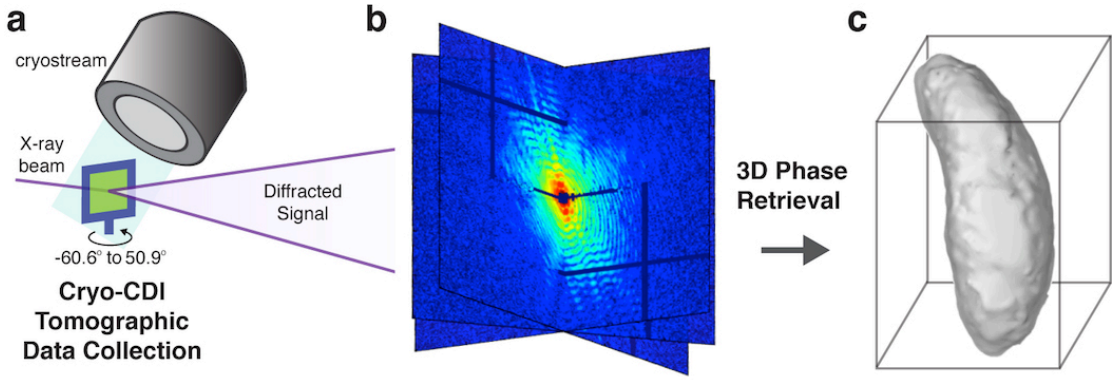

Figure 1: Schematic layout of a 3D cryo-CDI microscope. (**a**) A diagram shows a silicon-nitride-membrane containing frozen-hydrated cells on a single-tilt piezo-electric stage, bathed in a nitrogen gas cryosteam. A coherent X-ray beam impinges on a cell, from which a tilt series of 72 diffraction patterns was measured using a MAXIPIX detector in beamline ID10C at the ESRF (**b**). A 3D diffraction pattern was assembled from the 72 2D diffraction patterns, from which the 3D structure of the cell was iteratively reconstructed by using the oversampling smoothness (OSS) algorithm (**c**).

Ultrafast Imaging of Shocked Material Dynamics with X-ray Fee Electron Laser Pulses

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X-rays are particularly well suited for micron-scale materials studies due to their short wavelength and ability to penetrate bulk materials. Combining this ability with the intense ultrafast pulses from X-ray free electron lasers such as SLAC's Linac Coherent Light Source (LCLS) provides an ideal system for studying material response under extreme conditions such as impact, high load, or other non-equilibrium processes. Here we demonstrate this ground-breaking ability by showing sub-micron single-shot X-ray imaging of laser shocked materials performed at the Materials in Extreme Conditions (MEC) hutch at the LCLS in January 2014. We imaged the shock wave interactions with 10 micron voids in two single crystal materials: lithium floride (LiF) and the explosive pentaerythritol tetranitrate (PETN) with near 200 nm resolution. Shock wave interactions with voids in explosives are of particular interest due to the prevailing theory that the transition to detonation is caused by void collapse.

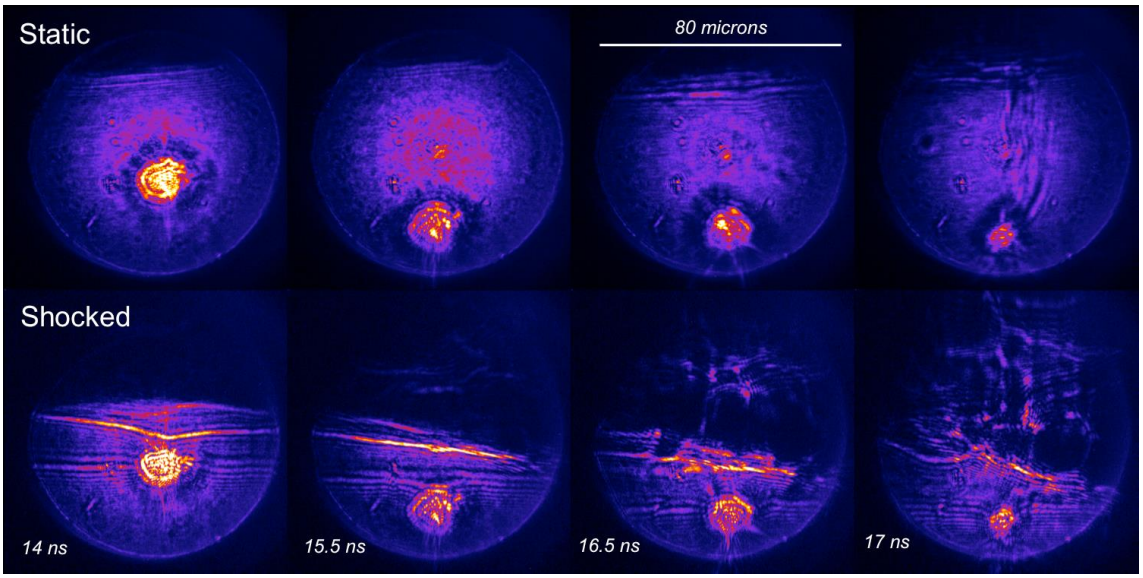

Figure 1: A montage of preliminary images comparing the static (top) and dynamic (bottom) single pulse images of four 10 micron diameter voids in 150 nm thick PETN at different delays with respect to the laser drive. The shock drive surface is at the top with the shock wave propagating towards the bottom. These images were taken on a scintillator couple CCD (FLI detector) with the detector placed 4.1 m behind the samples.

X-Ray Photon Correlation Spectroscopy At The APS With The Dectris EIGER Detector

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We have evaluated the suitability and performance of the Dectris EIGER detector for x-ray photon correlation spectroscopy (XPCS) as performed at beamline 8-ID-I at the APS. The EIGER detector is a newly-released photon counting bump-bonded area detector with 1,030 × 1,065 75-μm pixels that currently runs at a maximum sustained frame rate of 800 frames per second (fps) with 3,000 fps promised in the near future. With respect to other detectors currently used for XPCS at 8-ID-I, the EIGER promises high efficiency photon counting operation and, most significantly, considerably higher data rates than have previously been available (~ 3 gigapixels per second versus 0.1 gigapixels per second). Using the EIGER, we measured the ensemble-averaged small angle scattering from various reference samples and made a detailed comparison to measurements made using other detectors. We also measured the x-ray speckle contrast with and without focusing optics from static reference samples and found that the speckle contrast was easily observable even within beamline 8-ID-I's current layout that is not optimized for the larger pixels (75 μm versus 14, 20 or 30 μm) of the EIGER. Time correlation functions from dynamic reference samples were also obtained and agree well with previous work. In addition to the reference samples, we measured time correlation functions from flowing colloidal suspensions—measurements that have proven hard to obtain with other detectors. Lastly, we describe successful initial efforts to incorporate the high data rates from the EIGER detector into the XPCS high performance computing data pipeline at 8-ID.

Performance and measurements described in this abstract and data that will be presented on the poster were made with a detector on loan from Dectris. The object on loan is a prototype of the EIGER 1M detector system with limited functionality and performance as compared to the eventual production model. We acknowledge the support of the APS Detector Group in both securing the detector loan and in deploying the detector at 8-ID-I.

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ANL and LBNL 1k Frame Store CCD Detector Applied to XPCS at the APS

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We have commissioned the 1k Frame Store Fast CCD (1kFSCCD) detector for XPCS measurements at beamline 8-ID-I at the Advanced Photon Source (APS). The detector was developed as part of a collaboration between Argonne National Laboratory (ANL) and the Advanced Light Source (ALS) at Lawrence Berkeley National Laboratory (LBNL).

The 1kFSCCD detector uses a custom sensor (a CCD) and a custom readout integrated circuit both designed by LBNL. Backend electronics were designed by ANL. The active area of the sensor consists of 960 × 1,920 30-micron pixels in full mode or 960 × 960 pixels in frame store mode. The sensor output is digitized by 192 readouts each operating in parallel on 10 columns by 480 rows for fast readout. X-rays are detected directly by the silicon sensor; its thickness is approximately 300 microns providing highly efficient operation to 12 keV. The detector is currently being used at beamline 8-ID-I at the APS for XPCS measurements. In addition to the base modes described above, the detector can also be used with successively smaller regions of interest to achieve successively higher frame rates. One common mode of operation at beamline 8-ID-I is the 960 × 92 pixel mode that achieves a frame rate of 1,000 (partial) frames per second. A unique feature of the ANL version of the detector is a high bandwidth backplane connection within the detector control crate to a multi-node processor board that uses parallel programming with the Message Passing Interface (MPI) to perform on-the-fly compression of the 200 MB/s data stream generated by the 1kFSCCD detector allowing XPCS data acquisition sequences that can run at high frame rates for almost arbitrarily long periods of time.

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X-ray ptychographic imaging of human nuclei and chromosomes using reciprocal space up-sampling

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Our group is working on a BBSRC-funded project to study the structure of human chromosomes on the 30 nm scale by using X-ray diffraction. Here we demonstrate the application of Ptychography in biological imaging and show 2D ptychographic images of human nuclei and chromosomes with quantitative phase information, which can be used to determine the mass of the specimen. We also discuss subsampling of diffraction patterns in Ptychography to improve the performance of the iterative algorithm.

The Ptychography technique can yield aberration-free images with a wide field of view and in principle resolution limited by the wavelength of the X-rays [1,2]. The redundancy of information in Ptychography sets of diffraction patterns can be exploited to compensate for experimental limitations of the sampling: It was recently shown that Ptychography is not bound by the Nyquist-Shannon sampling theorem [3], and allows for reconstruction of the image even if it is violated [4]. We apply reciprocal-space up-sampling [5] to experimental data and obtain images of biological samples that exhibit weak absorption in the hard X-ray regime even from undersampled datasets. The phase information of the specimen obtained with this method is quantitative and directly related to the projected electron density. This information can be used to estimate the mass of the specimen in a unique way [6].

We show two-dimensional X-ray transmission images of unstained human nuclei and chromosomes, obtained hard X-ray ptychographic imaging at the Diamond Light Source. We also show mass estimations based on the quantitative phase information provided by ptychography.

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Nanosurveyor: A New Instrument for Nano-Tomography at the Advanced Light Source

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The Advanced Light Source has developed a tomographic microscope based on soft x-ray ptychography for the study of mesoscale materials [1]. The microscope utilizes an ultra-stable scanning mechanism with laser interferometer feedback for sample positioning and a fast frame rate charge-coupled device detector for soft x-ray diffraction measurements [2]. The microscope can achieve scan rates of greater than 20 Hz, including motor move, data readout and x-ray exposure, with a positioning accuracy of better than 2 nm RMS and achieved spatial resolution of better than 20 nm. A low numerical aperture condenser lens provides a well-defined x-ray probe on the sample with a 100 micrometer depth of field and 14 mm focal length. Such an optic is well suited for applications such as tomography and *in situ* measurements which require a long working distance. We have implemented a high performance data pipeline which enables real time ptychographic reconstructions on a distributed GPU-based architecture with a user friendly interface. This instrument, called Nanosurveyor, can achieve a spatial resolution nearly 20 times finer than the 300 nm x-ray spot in both two and three dimensions using 750 eV x-rays. Once moved to the new COSMIC beamline it will enable spectromicroscopy and tomography of materials with wavelength limited spatial resolution.

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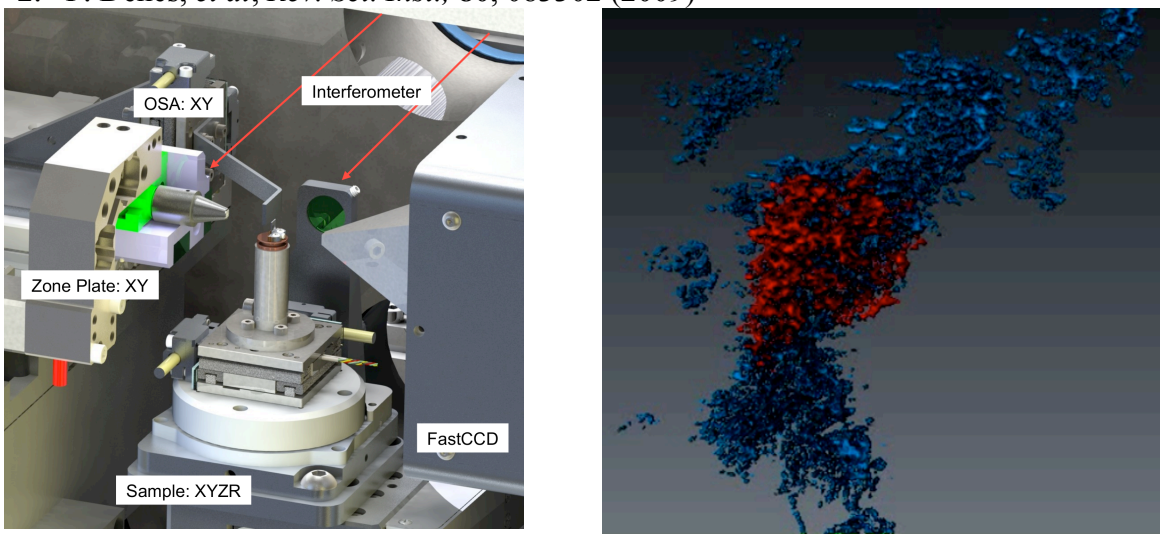

Figure 1: Nano-tomography by soft x-ray ptychography. (a) Nanosurveyor instrument showing the sample stage, zone plate scanning mechanism, order sorting aperture, interferometer and ALS fastCCD. (b) Tomographic volume rendering of porous Yttria stabilized Zirconia. The total volume is 3x3x3 micrometers and the voxel size is 9 nm. An average pore size of 72 nm is calculated from the reconstruction.

Soft X-ray Ptychography using holographic encoding

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We present high resolution images of biological test samples taken with ptychography, a scanning method that uses the overlap between two adjacent illumination positions to solve the phase problem for correctly oversampled diffraction patterns [1,2]. Diatom samples were imaged with ptychography at the U41-beamline at BESSY II at an energy of 400 eV in the soft x-ray regime. The experiment used an incident illumination from pinhole aperture and an additional small holographic reference hole in the pinhole mask. The addition of a reference hole allows the image from each scan position to be retrieved by Fourier Transform Holography (FTH) [3]. The position of the illumination can be determined from the images retrieved from FTH with very high accuracy, relaxing the need for an independent high resolution measurement of these positions, for example, via encoders. Good knowledge of the illumination positions is essential for ptychography and is a strong factor for obtaining high resolution images [4]. Detailed images of diatoms were obtained with 87 nm resolution using the positions calculated from the FTH images (fig. 1).

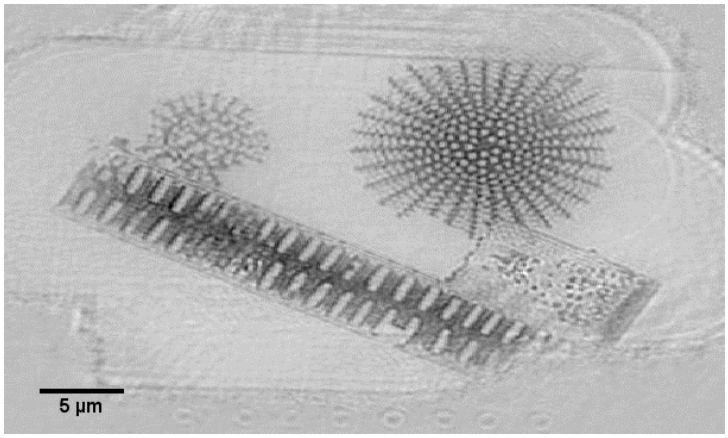

Fig 1. Phase image of Diatoms obtained from ptychography using positions retrieved from FTH images.

In the current set up a beamstop is used to block the high intensity of the central beam to best match the diffraction pattern intensity to the limited dynamic range of the 2D soft x-ray CCD detector. Missing information in the diffraction can, in some cases, cause the failure of the algorithms [5]. This problem has been solved both experimentally and using algorithms [6,7]. We present a moving beamstop in UHV that has vibrational modes designed to allow the intense low-q beam to be detected whilst being attenuated, with the ability to change the effective beamstop size and transmission in-situ. We compare the performance of ptychography with the central order blocked completely and with an attenuated beam.

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High-resolution multislice x-ray ptychography in combination with precession measurement

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X-ray ptychography, which is a scanning coherent X-ray diffraction imaging technique, can visualize both the transmission function of extended objects and the illumination function [1]. X-ray ptychography can be extended to three-dimensional imaging by means of tomography [2] and/or a multislice approach [3]. Multislice approach models the sample as a number of slices and the projection approximation is applied in a slice-by-slice manner. It is known that increasing the number of slices causes poor convergence in the phase retrieval calculation since the number of unknown variables will be much larger than that of independent equations. In order to overcome this problem, we propose the multislice X-ray ptychography in combination with the precession measurement. In this study, we carried out its feasibility study by computer simulation.

Figure 1(a) shows the second layer in the simulation model of four layers which is composed of "Osaka Univ.", "RIKEN", "SPring-8", and "SACLA" patterns. Figure 1(b) shows the image of the second layer reconstructed from the one ptychographical data set which was calculated at a single incident X-ray angle. The image includes not only the images of the other layers but also numerous artifacts. Figure 1(c) shows the image of the second layer reconstructed from the nine ptychographical data sets which were calculated at nine incident X-ray angles. Compared with Fig. 1(b), the image is dramatically improved. In this presentation, experimental results at SPring-8 will be also shown.

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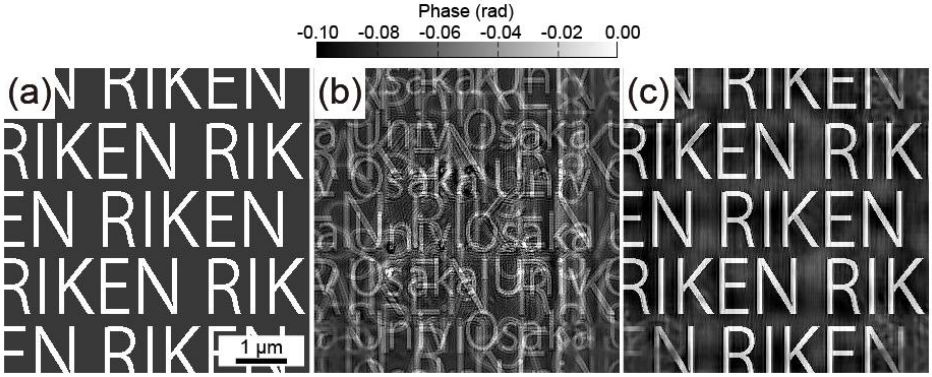

Figure 1: (a) Second layer in the simulation model. (b, c) Images of the second layer reconstructed from the ptychographical data set calculated at (b) a single incident angle and (c) nine incident angles.

Dynamics of silica particles in swollen rubber

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The addition of nanoparticles such as silica and carbon black to rubbery materials leads to the changes in bulk mechanical and viscoelastic properties. This reinforcement effect is indispensable for utilizing rubber materials as industrial products such as vehicle tires, while its physical picture has not been clarified. Filler nanoparticles form complex hierarchical structures; each stage of hierarchy shows specific responses to external forces, thereby resulting in the characteristic bulk viscoelastic properties. It is thus important to clarify the microscopic dynamics of nanoparticles in rubber. To elucidate the microscopic dynamics of nanoparticles in rubber, we have applied X-ray Photon Correlation Spectroscopy (XPCS) to rubber containing filler nanoparticles [1, 2]. The observed dynamics of nanoparticles shows temporal evolution (aging) and peculiar behaviors that are characterized by a relaxation showing compressed exponential behaviors. To shed light on the effects of network structure of rubber matrices on this peculiar dynamics of nanoparticles, we have applied XPCS to nanoparticles in swollen rubber with a different cross-link density.

XPCS experiments were performed at BL40XU, SPring-8. At BL40XU, high intensity quasi-monochromatic X-rays from helical undulator are utilized for producing quasi-coherent X-rays. Samples were swollen rubber containing spherical silica nanoparticles, the diameter of which was 1×10^2 nm. The results indicate that the irradiation of X-rays itself induces local rearrangement of nanoparticles depending on the cross-link density of rubber polymers. It is noteworthy that the azimuthally averaged scattering intensity profiles did not change and that the observed temporal evolution resembled what is commonly observed in the XPCS studies of complex fluids. In this presentation, we discuss the detailed effect of X-ray irradiation on the local rearrangement of silica in rubber.

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X-ray probes of local composition and structure near defects in InGaN multi-quantum wells

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Nitride semiconductors such as GaN, InN, AlN, and their alloys span a large range of direct band gaps, enabling many exciting applications such as high efficiency solid state lighting, high power switches, and semiconductor lasers. A lack of inexpensive, lattice matched substrates means that the epitaxial nitride thin films used in these applications have exceptionally large defect densities, which adversely affect device performance. [1] In light-emitting devices (LEDs) used in solid state lighting, high threading dislocation densities affect minority carrier concentration and recombination rates [2] within the InGaN multi-quantum-well (MQW) active regions of the devices, while compositional fluctuations are thought to localize carriers within the same region. [3] We report recent efforts to image composition variation, defects, and local structure in InGaN MQWs using focused coherent x-ray beams at Sector 26-ID of the Advanced Photon Source. Fluorescence mapping was used to determine composition fluctuations and identify v-defects, while simultaneous x-ray microdiffraction probed lattice rotations and strains. Coherent diffraction patterns from trench defects and v-defects found in these films were simulated using structural models and the structural sensitivity of Bragg ptychographic measurements to these defects was explored. [4] Preliminary reconstructions from experimental and simulated data will be discussed.

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Non-equilibrium structural dynamics of nanoparticles in $LiNi_{1/2}Mn_{3/2}O_4$ cathode under operando conditions

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Coherent x-rays are used to study non-equilibrium structural dynamics in lithium ion cathode (disordered $Li_{1-\delta}Ni_{1/2}Mn_{3/2}O_4$) during fast charge/discharge under operando conditions (see Figure 1a) [1]. We observe interference fringes due to finite size of the particles in the cathode material and are able to study transformation mechanisms on a single nanoparticle level (see Figure 1b). Our in-situ measurements in Bragg geometry reveal a hysteretic behavior of the structure upon cycling and we directly observe the interplay between different transformation mechanisms: solid solution and two-phase reactions (see Figure 1c). First attempts to study phase transitions using Bragg coherent diffraction imaging in battery materials [2] are also presented. Our study addresses the controversy of why two-phase materials show exemplary kinetics and opens new avenues to understand fundamental processes underlying charge transfer, which will be invaluable for developing the next generation battery materials.

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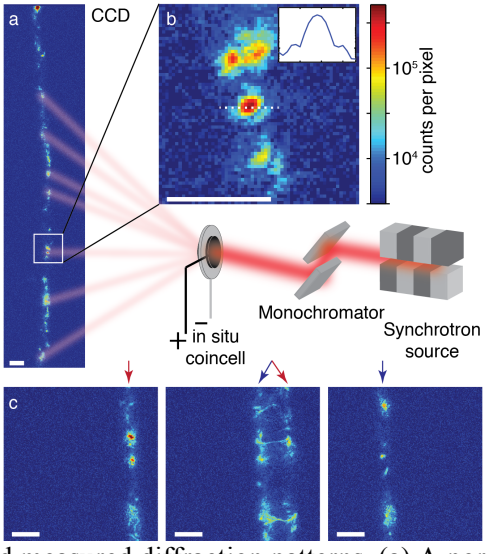

Figure 1. Experimental setup and measured diffraction patterns. (a) A portion of the Debye-Scherrer ring is recorded on a CCD positioned sideways at 2θ=17 degree. (b) An enlarged region of (a) shows fringes due to diffraction from a single cathode particle. (b, Inset) A line scan through the white dashed line in (b). (c) Measured diffraction patterns during discharge for lithium concentrations of 1-δ=0.12 (left pattern), 1-δ=0.7 (center pattern), 1-δ=0.9 (right pattern) as in $Li_{1-\delta}Ni_{1/2}Mn_{3/2}O_4$. The arrows in (c) indicate different structural phases. The scale bar in all images shows q=0.02 nm^{-1}.

2D AND 3D NANO-IMAGING USING NEAR-FIELD PTYCHOGRAPHY

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Inline holography or propagation-based phase-contrast is commonly used as a X-ray phase-contrast imaging method. Its implementation is especially simple since the phase signal is transferred to measured intensities through free-space propagation and no other image forming optical elements are required.[1,2] However, reconstruction of the sample's complex valued transmission function remains challenging for optically thick objects. In addition, a flat field correction is required which can fail in the case of strongly refracting samples.[3]

Here, we present a novel approach to simultaneously retrieve the sample's complex-valued transmission function and the incident illumination from Fresnel diffraction patterns by using the measurement diversity created by lateral translations of the sample with respect to a structured illumination.[5]

We show first results in 2D and 3D obtained with hard X-rays from synchrotron experiments and simulations on strongly and weakly refracting samples. We expect that our method will tremendously improve the robustness of X-ray phase nano-tomography.

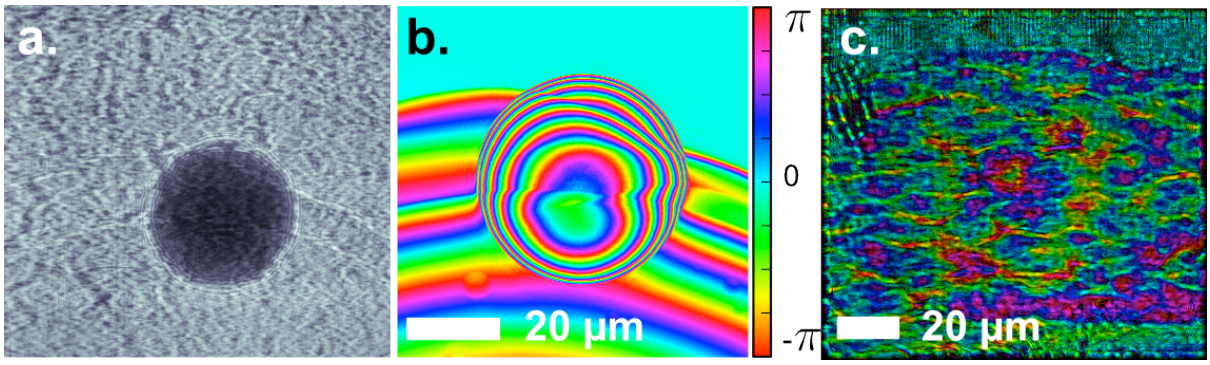

Figure 1: a, One of 16 diffraction patterns of a strongly absorbing(down to 5% transmission in the center) uranium sphere behind a structured illumination. **b,** reconstructed phase image of the uranium sphere. The sphere is seen to shift the phase up to 5 x 2 pi. **c,** reconstructed illumination function.

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Ptychographic overlap constraint errors and the limits of their numerical recovery using conjugate gradient descent methods

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Ptychographic coherent x-ray diffractive imaging is a form of scanning microscopy that does not require optics to image a sample. A series of scanned coherent diffraction patterns recorded from multiple overlapping illuminated regions on the sample are inverted numerically to retrieve its image. The technique recovers the phase lost by detecting the diffraction patterns by using experimentally known constraints, in this case the measured diffraction intensities and the assumed scan positions on the sample. The spatial resolution of the recovered image of the sample is limited by the angular extent over which the diffraction patterns are recorded and how well these constraints are known. Here, we explore how reconstruction quality degrades with uncertainties in the scan positions. We show experimentally that large errors in the assumed scan positions on the sample can be numerically determined and corrected using conjugate gradient descent methods. We also explore in simulations the limits, based on the signal to noise of the diffraction patterns and amount of overlap between adjacent scan positions, of just how large these errors can be and still be rendered tractable by this method.

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Challenges and capabilities of Coherent Bragg Imaging with nanoparticles

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Nanoparticles have found many applications in modern technology (catalysts, biosensors.…). Their properties and functionalities are dictated by the shape, the size, crystallinity, composition and chemical-state of the particle. A full characterization of individual particles remains a very challenging problem, due to limitations of current microscopy techniques. The emerging Coherent Bragg Imaging technique (CBI) [1,2] offers an effective way to evaluate the structural variation of a crystalline nanoparticle, which is difficult to be accessed by other methods. Up to now the best achieved resolution is ≈ 2 nm in a specific case [3], but is still not sufficient for characterizing nanoparticles with size below 50 nm. However new possibilities offered by 4th generations synchrotron sources open up new perspectives to investigate nano-particles down to 50-100 nm where surface effects are expected to be much more significant than larger nano-particles studied by CBI until now. These points are discussed with the new possibilities offered by HXN beamline at NSLS-II. The increase of incoming photons would permit to catch appreciable signal from small diffracting volume and access larger area in reciprocal space resulting in increased resolution of the reconstructions in direct space. The achievable resolutions for different nano-particle type and size (i.e. Au, Ag, Pt, Pd or Fe2O3) will be evaluated.

Waiting for experimental data to go further in testing sensitivity of CBI and phase retrieval, some of the systematic errors (interpolation, FFT, phase retrieval algorithms…) are quantified through simulation. A comparison between retrieved and initial synthetic particles is performed to calculate error bar as a function of studied parameter (i.e. photon counts/ particle size…). In addition to idealized bulk-like nanoparticles, structural models were also generated using first principle density functional theory (<10^3 atoms) and classical embedded atom method potentials (10^3-10^6 atoms) to provide realistic atomic positions of the particles, in particular to include influence from e.g. surface relaxation, disorder... The case of highly distorted or pseudo-amorphous particles (where classical approaches still have to be tested/modified) will be emphasized.

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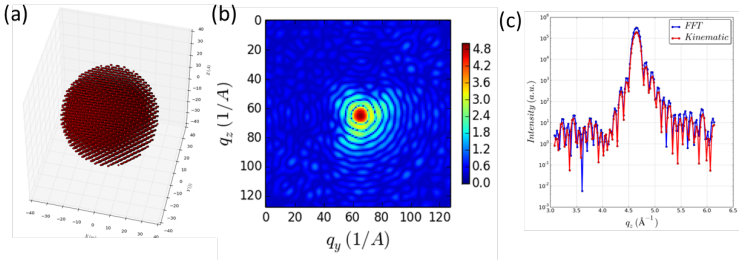

Figure 1: (a) Example of synthetic spherical 6 nm Au nano-particle relaxed by DFT [4] (b) Reciprocal space 2D slice extracted from the 3D maps of (333) Bragg peak. (c) FFT and kinematic sum comparison.

Challenges and capabilities of Coherent Bragg Imaging with polycrystalline thin film during mechanical loading

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Supported polycrystalline thin films are ubiquitous in many applications ranging from micro-devices to hard coatings. The grain size generally scales with the film thickness and both inter- and intra-grain strain heterogeneities are expected. The origins and magnitudes of these strain heterogeneities are of great interest in technology because many fabrication and reliability problems are stress related. But measuring local strains in sub-micron size grains remains a real experimental challenge.

An original multi-scale strain determination approach is proposed to address this issue [1-3]. Both micro and C**oherent X-ray diffraction** are used to study a model {111} textured Au thin film during thermal cycles. Individual average strain in few tens of grains (inter-grain heterogeneities) is monitored with micro-diffraction. Intra-grain strain (≈ 20 nm resolution) is evaluated thanks to coherent diffraction and phase retrieval during a similar cycle. Potentialities and limitations of these approaches to study polycrystalline behaviours are discussed.

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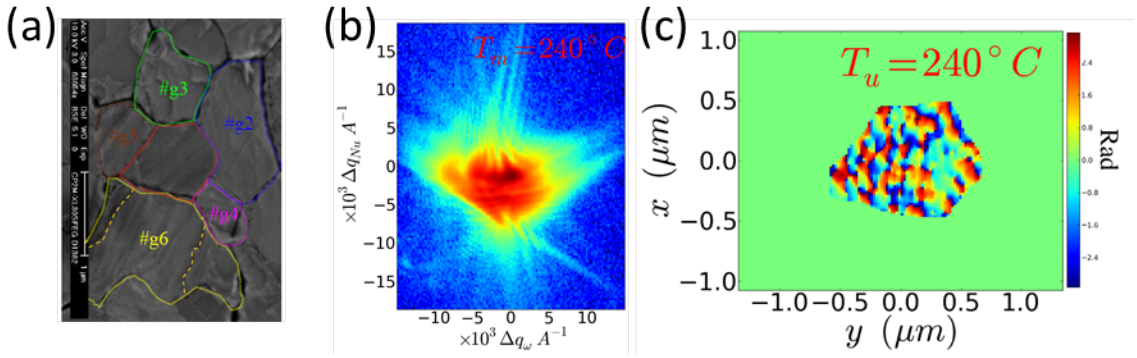

Figure 1: (a) SEM of study grain from an Au polycrystalline thin film (b) (qx,qy) slice of 3D (111) Bragg peak (recorded in symmetric coplanar geometry) (c) Retrieved uz displacement field (Φ=G.u) at T=240°C.

A novel real-time beam monitor for coherent and ultra-fast imaging

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The interpretation of images arising from coherent imaging microscopy techniques relies heavily on the understanding of the illumination of the sample. This is particularly important at 4th generation sources, such as the Linac Coherent Light Source (LCLS), due to the stochastic, pulsed nature of the x-ray beam delivered to the sample. At the coherent x-ray imaging instrument (CXI) of LCLS—which operates in the 4-20keV range—the peak x-ray power can exceed 10^{20} W/cm^2, which is sufficient to alter the structure of any sample in a single pulse and calls into question the accuracy of using time-averaged beam reconstructions for data interpretation. Here, we describe how the transport of intensity equation can be used in conjunction with the prototype of a minimally disruptive, real-time wave-front monitor for 3rd and 4th generation x-ray sources.

Opportunities and challenges with high-energy coherent X-rays

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High brilliance third generation synchrotron sources allowed the development of new scattering techniques exploiting coherent hard X-ray beams such as X-ray Photon Correlation Spectroscopy (XPCS) [1] and Coherent X-ray Diffractive Imaging (CXDI) [2]. XPCS has been applied to study dynamics in colloidal suspension [3, 4], liquid surface fluctuations [5], atomic motion in glasses [6] and alloys [7]… CXDI is progressively moving from demonstration experiments [2, 8] to imaging of relevant inorganic and biological specimens in 3D [9, 10]. Yet, many systems remain unexplored due to the radiation damage or strong absorption since most of these experiments are performed with an incident photon energy E below 10 keV. High energies open new possibilities for these techniques, in particular for XPCS. The E^{-2} loss of coherent flux can be in some cases compensated by larger sample volumes thanks to the E^{-3} dependence of the absorption, especially in small-angle geometry. In addition, the high penetration power of 20-30 keV X-rays allows the study of systems in bulky environments, e.g. at liquid-liquid or liquid-solid interfaces, at high pressure in diamond anvil cells, etc. The first demonstration of high energy XPCS [11] showed a great potential that was limited by the low coherent X-ray flux. With the progress in accelerator technology the future storage rings promises a 30 fold increase in brilliance. This will provide a coherent flux at 20-30 keV few times higher than the present values at 7-10 keV.

Here we show recent results with coherent high energy X-rays at the renewed ESRF beamline ID10. We compare XPCS measurements at 8 keV with 21 keV in small-angle and wide-angle geometries. To take full advantage of high energy, coherent X-rays efficient detectors are needed like the new MAXIPIX pixel detector with CdTe chips. We also show that CXDI can benefit from high energies only with the development of detectors with small pixel size.

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Coherent X-ray diffractive imaging at the ESRF beamline ID10: possibilities and challenges

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Coherent X-ray diffractive imaging (CXDI) is a novel scattering technique [1,2] that exploits the unprecedented degree of coherence of modern synchrotron sources. It has potential for high resolution imaging of isolated microscopic objects beyond the values achieved with X-ray lenses and represents an interesting tool to bridge the gap between high resolution electron and visible light microscopy. The image in the real space is obtained by applying phase retrieval algorithm to the diffraction pattern measured with sufficient oversampling. Because of the high penetration power of the X-rays the imaging of thick object (<10μm) without sectioning is possible in 3D [3,4,5].

Here we report the current status of the CXDI at the ESRF beamline ID10. We discuss the possibilities and challenges of CXDI by presenting examples of 2D reconstruction of *Deinococcus radiodurans* (DR) bacteria (Fig. 1 (left)) and 3D reconstruction of a porous Si cluster (Fig. 1 (right)). The high quality of the reconstructed images reveals DR's cellular structure and individual Si nano-crystals with unprecedented detail. The improvement in the biological sample preparation and the development of large 2D (pixel) detectors are the key elements for achieving the full potential of the technique.

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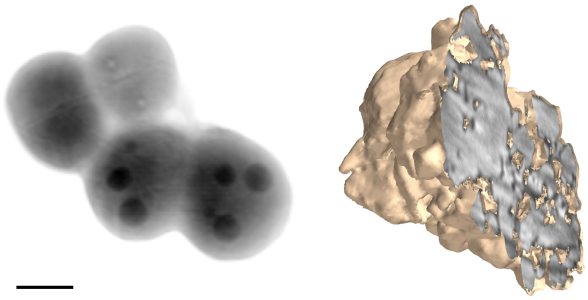

Figure 1: Reconstructed 2D image of bacteria *Deinococcus radiodurans* (left) and a cut through a 3D image of a porous cluster of 100-200nm Si crystals (right). The scale bar in the left image corresponds to 1 μm.

Ptychographic coherent diffraction imaging of polystyrene colloidal crystals in the vicinity of melting transition

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3D assemblies of colloidal particles of sub-micron size represent a new class of materials with potential applications in photonics and optoelectronics [1]. Photonic properties of colloidal crystals can be modified by sintering and annealing the material at elevated temperatures [2]. Of special interest is the temperature range in the vicinity of crystal melting, when the long-range ordering vanishes, which has important technological aspects regarding the performance of a photonic device.

Among the various techniques used to assess the structure of colloidal systems the X-ray diffraction at third generation synchrotron sources enables *in situ* non-destructive studies of colloidal crystals [3, 4]. Further possibilities of *ab initio* image reconstruction are offered by the coherent diffraction imaging technique (CDI) based on the coherent illumination of a sample and measuring the diffraction patterns in the far diffraction field [5]. Missing phases of scattered waves can be iteratively retrieved by computational algorithm implying constrains in direct and reciprocal space. Considerable enhancement of phase retrieval convergence and larger field of view can be achieved using ptychographic coherent diffraction imaging (PCDI) based on collecting multiple diffraction patterns from overlapping positions on a sample [7].

In this work the PCDI technique has been applied to directly monitor the structural evolution of colloidal crystals upon heating. We performed *in situ* PCDI experiments using a microfocused beam at the coherence beamline P10 of PETRA III synchrotron (DESY Hamburg). Highly ordered crystalline domains in a colloidal crystal made of spherical polystyrene particles were studied in transmission geometry. The PCDI data were collected over a square mesh on the same sample area at each temperature during incremental heating up to the melting point (T ~ 380 K). The reconstructed ptychographic images revealed several stages of structural evolution in the colloidal crystal upon heating treatment. The reported PCDI study represents a novel approach to *in situ* monitoring of structural changes in colloidal crystals.

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